

An inventory of gaseous and primary aerosol emissions in Asia in the year 2000

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Abstract. An inventory of air pollutant emissions in Asia in the year 2000 is developed to support atmospheric modeling and analysis of observations taken during the TRACE-P experiment funded by the National Aeronautics and Space Administration (NASA) and the ACE-Asia experiment funded by the National Science Foundation (NSF) and the National Oceanic and Atmospheric Administration (NOAA). Emissions are estimated for all major contributing sources, including biomass burning, in 64 regions of Asia. We estimate total Asian emissions as follows: 34.3 Tg SO₂, 26.8 Tg NO_x, 9870 Tg CO₂, 279 Tg CO, 107 Tg CH₄, 52.2 Tg NMVOC, 2.54 Tg black carbon (BC), 10.4 Tg organic carbon (OC), and 27.5 Tg NH₃. In addition, NMVOC are speciated into 19 sub-categories according to functional groups and reactivity. Thus, we are able to identify the major source regions and types for many of the significant gaseous and particle emissions that influence pollutant concentrations in the vicinity of the TRACE-P and ACE-Asia field measurements. Emissions in China dominate the signature of pollutant concentrations in this region, so special emphasis has been placed on the development of emission estimates for China. China's emissions are determined to be as follows: 20.4 Tg SO₂, 11.4 Tg NO_x, 3820 Tg CO₂, 116 Tg CO, 38.4 Tg CH₄, 17.4 Tg NMVOC, 1.05 Tg BC, 3.4 Tg OC, and 13.6 Tg NH₃. Emissions are gridded at a variety of spatial resolutions from 1° × 1° to 30 sec × 30 sec, using the exact locations of large point sources and surrogate GIS distributions of urban and rural population, road networks, land-cover, ship lanes, etc. The gridded emission estimates have been used as inputs to atmospheric simulation models and have proven to be generally robust in comparison with field observations. Monthly emission estimates for China are developed for each species to aid TRACE-P and ACE-Asia data interpretation. During the observation period of March/April, emissions are roughly at their average values (one-twelfth of annual).

Uncertainties in the emission estimates, measured as 95% confidence intervals, range from a low of $\pm 16\%$ for SO_2 to a high of $\pm 450\%$ for OC.

INDEX TERMS: 0305 Aerosols and particles; 0322 Constituent sources and sinks; 0345 Pollution—urban and regional; 0365 Troposphere—composition and chemistry

1. Introduction

A good understanding of air pollution concentrations and fluxes at regional scale requires the integration of observations from field campaigns with results from regional or global atmospheric models over the same domains of time and space. To exercise the atmospheric models requires a coordinated set of emission inputs of all necessary species. It is important that these emission fields correctly reflect the spatial and temporal emission profiles of sources that were operating during the time period of the field campaigns, otherwise good agreement between models and experiment cannot be expected. Knowledge of source strengths and locations is also a valuable aid for interpreting observations and model results and ultimately choosing appropriate mitigation strategies.

For these reasons we have developed an emission inventory for Asia for the year 2000 to contribute to the NASA TRACE-P (Transport and Chemical Evolution over the Pacific) Mission and ACE-Asia (Asian Pacific Regional Aerosol Characterization Experiment) funded by NSF and NOAA. This is the first time that Asian emissions have been studied in a comprehensive and consistent way for a recent year. Though there are considerable uncertainties associated with some of the emissions values, due to lack of national statistics in many countries and lack of knowledge about the performance of certain kinds of emitters, we nevertheless believe that the confidence level for many species is high and that the inventory will be valuable in gaining an understanding of the formation and fate of regional air pollutants in Asia.

The domain stretches from Pakistan in the West to Japan in the East, and from Indonesia in the South to Mongolia in the North. Figure 1 shows the countries and sub-

regions that are part of this inventory. International shipping lanes are included, located according to the method of *Streets et al.* [2000b]. In order to focus on the needs of the two missions, the following priorities were given to emissions estimation: first, China; second, the rest of East Asia; third, Southeast Asia; and fourth, South Asia. Emissions are estimated for each country in the region and for each province of China. Often in this paper we emphasize emissions from China and the rest of East Asia, because they dominate the pollution signature along the TRACE-P and ACE-Asia flight paths (in the South China Sea, East China Sea, Yellow Sea, the Sea of Japan, and the western Pacific Ocean).

Figure 2 shows the general methodology for this activity. Emissions are estimated for nine major chemical species: SO₂, NO_x, CO₂, CO, CH₄, non-methane volatile organic compounds (NMVOC), submicron black carbon aerosol (BC), submicron organic carbon aerosol (OC), and NH₃. In addition, NMVOC emissions are speciated into 19 sub-categories based on chemical reactivity and functional groups. Note that primary PM₁₀ and PM_{2.5} emissions are not presented in this paper but have been independently estimated by this team, in coordination with the BC and OC emissions. Emissions are calculated for each source type believed to contribute significantly to the emissions of a particular chemical species. As examples, ten transportation categories are examined for all species and 82 emitting categories are compiled for NMVOC emissions. For energy use we often rely on the RAINS-Asia computer simulation model, as described below, supplemented with non-energy activity data for such species as NMVOC, CH₄, and NH₃. Local regulations limiting the emissions of air pollutants are included in the model through the application of specific (required) abatement technologies.

Emissions presented in this paper are for anthropogenic sources only. We do not include such natural sources as volcanic SO₂, biogenic NMVOC, CH₄ from wetlands, or mineral dust; these are calculated on-line and added into the total gridded emissions as an integral part of the modeling process [*Carmichael et al. (a)*, this issue]. Biomass burning *is* included in this inventory because it is largely caused by human activity. (Even in remote areas it is believed that lightning-induced fires are a minor cause of burning.) Biomass burning is divided into three major categories: forest burning, savanna/grassland burning, and the burning of crop residues in the field after harvest. The reader should note that we distinguish carefully between this kind of “open” biomass burning and the combustion of biofuels (wood, crop residues, dung, etc.) in stoves, cookers, and heaters, which is considered to be an energy source alongside fossil-fuel combustion. The detailed emission calculations are aggregated into seven primary source categories: industry, residential, transportation, power generation, agriculture, biomass burning, and other. It is at this aggregated level that summaries of emissions are presented later in this paper.

Emission estimates are specifically for the year 2000, so as to incorporate many of the fundamental changes that occurred in Asia in the late 1990s. However, when this project began in late 2000—in order to provide pre-mission modeling support to the field experiments that took place in March and April 2001—very little statistical data had been released for the year 2000 by Asian countries. Thus, our approach consists of a mixture of actual year-2000 data, model forecasts for 2000 from a 1995 base, and trend extrapolations from emission estimates for the late 1990s.

Section 3.1 presents the results for each species and compares the results with other literature estimates. This section also compares and contrasts emissions among

species, sources, and countries. A separate discussion of the sources of individual organic chemical species and the differences among countries is presented in Section 3.2.

Emissions are initially calculated for an annual time period. However, it is recognized that there is considerable seasonal variation for some species, associated with such activities as the burning of fossil fuels for home heating in winter and the temperature dependence of releases of NH_3 from fertilizer application and CH_4 from manure. Biomass burning clearly has a very high degree of seasonality, determined by local agricultural practices and meteorological conditions. Such seasonality is important in preparing emissions for comparison with time-specific field experiments. For this reason we have apportioned annual emissions to daily and monthly emissions using a variety of techniques discussed in Section 3.3. The techniques used to develop biomass burning estimates are elaborate and based on national surveys of fires, AVHRR satellite fire counts, and TOMS AI data; they are described in two separate papers [Woo *et al.*, this issue; Yarber *et al.*, manuscript in preparation, 2002]. Efforts concentrated on realistically apportioning annual emissions to the months of March and April, which correspond to the timing of the TRACE-P and ACE-Asia field campaigns.

As Figure 2 shows, anthropogenic emissions are initially calculated for 64 regions of Asia. In this project we do not separately report emissions from the 22 major cities of Asia that are included in the model. Regional emissions are then gridded at a variety of spatial resolutions for input to the atmospheric simulation models, ranging from $1^\circ \times 1^\circ$ for most regional model applications down to $30 \text{ sec} \times 30 \text{ sec}$ resolution for urban-scale studies. Each type of emission source is represented by a different spatially resolved surrogate parameter, using such data sets as urban and rural population, road networks,

land cover, and ship lanes. Approximately 355 large Asian power plants are separately identified in the RAINS-Asia model and located according to their latitude/longitude coordinates. For input to the atmospheric models, gridded representations of anthropogenic area and point sources are developed separately, and natural sources are added at the end. Thus, all atmospheric releases are ultimately accounted for, such that comparison with observations can be meaningfully considered. The development of the gridded emission fields, the sources of surrogate GIS data that were used, and some examples are presented in Section 3.4.

An uncertainty analysis is presented, focusing on the combined uncertainties of activity levels and emission factors for each species in each of six prototypical regions. The uncertainty results are presented in Section 3.5. Finally, in Section 3.6, we present comparisons between modeled pollutant concentrations derived using this emission inventory and observed concentrations during the TRACE-P and ACE-Asia missions and at other ground stations. The findings are used to qualitatively assess the level of confidence that can be placed in the inventory values and, in some cases, their apparent shortcomings. All regional and gridded emissions data sets can be examined and downloaded from our emissions website at the University of Iowa. The url address is:

http://www.cgrer.uiowa.edu/EMISSION_DATA/index_16.htm.

2. Methodology

The estimation of emissions from such a wide variety of source types, species, and regions cannot be described in complete detail due to space limitations. However, we can give a general overview of the methods, data, and data sources for the key elements

of the emission calculations. The general approach used by us to estimate emissions has been described in *Klimont et al.* [2002]. The emissions of a particular species are estimated as a product of the activity rate, the unabated emission factor, and the removal efficiency of any applied emission abatement technologies, using the following equation:

$$E_{j,k} = \sum_l \sum_m \sum_n A_{j,k,l,m} ef_{j,k,l,m} (1 - \eta_{j,l,m,n} \alpha_{j,k,l,m,n}) X_{j,k,l,m,n} \quad (1)$$

where,

j,k,l,m,n	= species, region, sector, fuel/activity type, abatement technology;
E	= emissions;
A	= activity rate;
ef	= unabated emission factor;
η	= removal efficiency of abatement technology n ;
α	= maximum application rate of abatement technology n ; and
X	= actual application rate of abatement technology n ; note that the set of abatement technologies includes a ‘no-control’ case, such that $\sum_n X = 1$.

The parameter $\alpha_{j,k,l,m,n}$ is used only for NMVOC in this inventory to allow for the fact that emissions are generated by a large number of inhomogeneous sources, and consequently some control options apply only to a fraction of the total activity. Throughout most of developing Asia, there is little abatement technology in place, such that $(1 - \eta_{j,l,m,n} \alpha_{j,k,l,m,n}) X_{j,k,l,m,n} = 1$, and emissions become simply a product of activity rates and emission factors. Abatement technologies must be considered for many species and sources in the developed countries of Asia (Japan in particular); however, in the rest of Asia it is generally only large sources of PM and SO₂ in the industry and power generation sectors for which abatement technologies must be considered. We discuss below the development of activity rates and emission factors. Additional detailed information on emissions by sub-sectors and emission factors can be found on the web site.

2.1. Activity Rates

Because of the need to generate a comprehensive picture of energy use by fuel type, sector, and Asian region, we often used the year-2000 forecasts of the RAINS-Asia simulation model from a 1995 base [Foell *et al.*, 1995; Streets *et al.*, 1999; Shah *et al.*, 2000]. One important exception is that of China, for which fundamental structural changes and a slowdown in the Chinese economy led to a reduction in coal use after 1996-97 [Sinton and Fridley, 2000; Streets *et al.*, 2001b], rather than the increase projected by the RAINS-Asia model; for China we therefore developed our own estimates of fuel use by sector and province based on the work of Sinton and Fridley [2000]. Table 1 presents the basic information on fossil-fuel and biofuel energy use in the year 2000 by country, sector, and fuel type that was used to develop combustion-related emissions. The quantities of biomass burned are also included in Table 1, the derivation of which is described in a separate paper [Yarber *et al.*, manuscript in preparation, 2002]. Table 2 identifies the other, non-combustion source types that are included in the emission calculations, though we do not present the activity rates for these source types.

A separate approach was taken for transportation emissions, because of the need to consider the variety of vehicles in use in Asia and their widely differing emission rates. Our approach was to estimate emissions as the product of numbers of vehicles in a country, annual vehicle km traveled, and emission rate in g km^{-1} of pollutant emitted. The numbers of vehicles and distances traveled were obtained where possible from World Road Statistics 2000 [IRF, 2000] and World Motor Vehicle Data [AAAA, 1998], inferred for some countries and vehicle types from national data sources and extrapolations. Table 3 shows the assumed numbers of each of ten types of vehicles in each country.

The methods for determining activity levels for NMVOC and BC are the same as in previous analyses [*Klimont et al.*, 2001, 2002; *Streets et al.*, 2001c]. Activity levels for agricultural and related data needed for estimating CH₄ and NH₃ emissions were derived from national statistics wherever possible (e.g., *NBS* [2000]) and international statistics (e.g., *IFA* [1998]; *FAO* [2000]; *OECD* [1995]). Often, data for 2000 were not available and were extrapolated from recent statistical information for 1998 and 1999 or derived from short-term forecasts [*OECD*, 1999].

2.2. Emission Factors

Emission factors were developed from a wide range of sources. Table 4 summarizes the ranges of emission factors that were used for fossil-fuel and biofuel combustion in different sectors. The ranges presented cover different countries or provinces; typically, the low end of the ranges applies to the most developed regions, like Japan or Hong Kong, and the upper end of the ranges applies to less-developed regions, like Laos or Xinjiang Province.

Emission factors for vehicles were derived using the MOBILE5 model, as described for China in *Fu et al.* [2001]. Emission factors were determined for each of the ten vehicle categories for six prototypical countries—China; India; Japan; Republic of Korea; Taiwan, China; and Thailand. Other countries were then assigned the same emission factors as the modeled country they most closely resembled. Emission factors for open biomass burning were taken from *Andreae and Merlet* [2001] and are shown in Table 5. Emission factors for CH₄ essentially followed IPCC methodology [*IPCC*, 1997], with special use of the China Country Study [*Tsinghua University and others*, 1999].

Emission factors for NH_3 are based on *Bouwman et al.* [1997], but with livestock emissions adjusted for region-specific production efficiency of milk and meat and fertilizer emissions adjusted for differences in nitrogen losses between temperate and tropical regions [*Klimont et al.*, 2001]. For some source types that we believe are similar in Asia and the West (large power plants, for example), we used USEPA AP-42 emission factors [USEPA, 2002]; these values were sometimes adjusted to reflect poorer performance of Asian technologies (see Section 3.1.4). Emission factors and particulate collection efficiencies for BC are described in *Streets et al.* [2001c], and an overview of our assumptions about BC and OC emission factors will be reported elsewhere in the context of a global inventory (T.C. Bond *et al.*, manuscript in preparation, 2002).

In selecting emission factors to be used in the development of this inventory, we were struck by the wide variation in literature estimates. One simple illustration of this variation can be obtained by comparing CO emissions from the stove tests of *Zhang et al.* [1999] and *Venkataraman and Rao* [2001]. The former, testing 15 wood-fired stove types in India and China, obtained a mean estimate of $78 \text{ g kg}^{-1} \text{ CO}$, with values in the range of $42\text{--}170 \text{ g kg}^{-1}$. For four stoves fired with dung cake the values obtained by *Zhang et al.* [1999] were $43 \text{ (30--61) g kg}^{-1} \text{ CO}$. *Venkataraman and Rao* [2001], on the other hand, report measured values of $11\text{--}12 \text{ g kg}^{-1} \text{ CO}$ for four Indian wood-fired stoves and $14\text{--}29 \text{ g kg}^{-1} \text{ CO}$ for four Indian stoves fired with dung cake. Such differences point out the very great and urgent need for comprehensive and rigorous testing of emission sources in the developing world—and also partly explain why our uncertainty estimates (Section 3.5) are so large for species like CO, BC, and OC, the emissions of which are dominated by small combustion sources.

3. Results

3.1. Asian and National Emission Estimates

This section presents national and regional summaries of emissions of each of the nine major species studied and places them in the context of other emissions values reported in the literature. It also identifies the major source types contributing to the emissions of each species. Table 6 summarizes national emissions of each species.

3.1.1. Sulfur dioxide. Of all the pollutants emitted in Asia, SO₂ is best known. Prompted by concerns over high ambient SO₂ concentrations in Asian cities and the threat of acid rain, Japanese researchers commenced studies of SO₂ emissions in Asia more than ten years ago [*Fujita et al.*, 1991; *Kato and Akimoto*, 1992; *Akimoto and Narita*, 1994]. SO₂ emissions in Asia rose steadily to a peak of 39.9 Tg in 1996 [*Streets et al.*, 2000a], as ever-increasing quantities of fossil fuels were used to power rapidly growing industrial economies. Since 1996, however, SO₂ emissions have slowly declined, due to a combination of economic slowdown, environmental regulations, and the fundamental restructuring of the Chinese industrial economy [*Sinton and Fridley*, 2000]. We estimate that Asian SO₂ emissions in 2000 were 34.3 Tg, which is in good agreement with the time trend of *Streets et al.* [2000a] through 1997. The power generation sector, dominated by coal-fired power plants in China, produced 45% of total Asian SO₂ emissions in 2000, and the industrial sector produced 36%.

Emissions of SO₂ in China in 2000 are estimated to be 20.4 Tg. This is in excellent agreement with the official Chinese estimate of 19.95 Tg [*SEPA*, 2000]. Though we do not know the details of SEPA's calculation, it is likely that it does not include some contributing sources that we do include, such as domestic biofuel

combustion (450 Gg), biomass burning (83 Gg), and Hong Kong sources (76 Gg)—so our estimate may actually be in even better agreement. An estimate for 1995 of 25.2 Tg [Streets and Waldhoff, 2000] is higher than the official SEPA value of 23.7 Tg, for reasons discussed in the text of that paper. We estimate emissions of 5.54 Tg in India in 2000. This can be compared with recent estimates of 4.35 Tg for 1996-1997 by Reddy and Venkataraman [2002a,b] and 4.64 Tg for 1995 [Garg *et al.*, 2001b]. India's emissions of SO₂ have grown unabated throughout the 1990s, at an annual rate of about 5.5% according to Garg *et al.* [2001b], so our estimate is quite consistent. Our estimates for other countries in Asia—such as 801 Gg for Japan; 829 Gg for the Republic of Korea; 376 Gg for Taiwan, China; and 961 Gg for Thailand—can be seen to be reasonable from both the long-term modeled trends and the official estimates through 1997 presented in Streets *et al.* [2000a]. Additional support for the validity of our SO₂ emission estimates is provided by the good agreement for SO₂ and SO₄ concentrations and wet sulfate deposition between values simulated by eight long-range transport models using our emission estimates and observations at 18 surface sites in East Asia [Carmichael *et al.*, 2002a]. A detailed treatment of long-term SO₂ emission and deposition trends can be found in Carmichael *et al.* [2002b].

3.1.2. Nitrogen oxides. We estimate that total NO_x emissions in Asia were 26.8 Tg in 2000 (all values herein calculated as NO₂). There are no other estimates of recent NO_x emissions in Asia against which to compare. Akimoto and Narita [1994] estimated Asian emissions in 1987 to be 15.5 Tg, van Aardenne *et al.* [1999] estimated emissions in 1990 to be 19.2 Tg with a projection for the year 2000 of 31.9 Tg, and the 1990 value for Asia from the global inventories of Olivier *et al.* [1996] is 23.1 Tg. All

researchers agree that the rapid growth of the transportation sector in Asia has caused NO_x emissions to climb steeply. Therefore, our numbers seem reasonable continent-wide and consistent with historical NO_x emission trends and other earlier Asian estimates [Streets *et al.*, 2001a]. The transportation sector has the highest share of Asian NO_x emissions (37%), followed by power generation (27%) and industry (18%).

For China, we calculate that emissions in 2000 were 11.3 Tg. Power generation has the largest contribution in China (39%), with industry and transportation both contributing about 25%. Hao *et al.* [2002] recently published estimates of NO_x emissions from commercial energy use in China in the period 1995-1998. A declining trend since 1996 is presented by Hao *et al.* [2002] (12.03 Tg in 1996, 11.66 Tg in 1997, and 11.18 Tg in 1998), consistent with the declines in emissions of other species in China reported elsewhere in this paper. Because Hao *et al.* [2002] only consider commercial energy, our value can be expected to be higher because of the inclusion of biofuel consumption and biomass burning. We also show a lower share of industrial emissions, consistent with reduced industrial coal use (Sinton and Fridley [2000]), and a higher share of transportation emissions, due to the rapid growth in vehicle ownership in China in the latter part of the 1990s. The value of 11.99 Tg for 1995 reported by Streets and Waldhoff [2000] is consistent with the value of Hao *et al.* [2002] for 1996, but higher than that reported by Klimont *et al.* [2001] for 1995, i.e., 9.7 Tg. Wang *et al.* [1996] reported that NO_x emissions in China in 1990 were 8.4 Tg, showing just how fast emissions grew during the early to mid-1990s.

For much of the rest of East Asia (Japan; Republic of Korea; and Taiwan, China) our estimates are in good agreement with government statistical reports. For the rest of

Asia, however, there are few estimates of NO_x emissions against which to compare. *Garg et al.* [2001b] report an estimate of 3.46 Tg for India's NO_x emissions in 1995, growing at an annual rate of about 5.5%. This value tallies well with our year-2000 estimate of 4.59 Tg. *Garg et al.* [2001b] calculate that in 1995 the power and transportation sectors both contributed about 28% to total NO_x emissions in India; our work suggests that by 2000 the share of the transportation sector had risen to 34%, while power generation had dropped to 26%.

3.1.3. Carbon dioxide. Though this study is not intended to support global climate change studies, atmospheric scientists are interested in local CO₂ concentrations and their relationships to concentrations of other carbon compounds like CO and CH₄. Our estimate for CO₂ emissions in Asia in the year 2000 is 9870 Tg. This estimate consists of all direct releases, including biomass burning, biofuel combustion, and releases from cement production, but it does not include any uptake from growing vegetation. Some researchers do not include vegetation burning in their CO₂ accounting, arguing that the resource is renewable and the released CO₂ is taken up by re-growth of the vegetation in later time periods. We prefer to separate the accounting of CO₂ release from that of CO₂ uptake. We have shown [*Streets and Waldhoff*, 1999] that throughout Asia vegetation combustion is so inefficient that it releases large quantities of non-CO₂ gases and particles (such as CO, CH₄, NMVOC, and carbonaceous aerosols), which may not be returned to the vegetative ecosystem as usable carbon for a long time. This can amount to 10-15% of the carbon in the fuel. In addition, much harvesting and burning of vegetation in Asia is non-sustainable. We leave it to others to balance this released CO₂ with carbon uptake in Asia.

As part of its global emissions inventory, the IPCC estimates 7440 Tg for anthropogenic CO₂ releases in Asia in 2000 [Nakicenovic *et al.*, 2000], compared with our value of 8730 Tg (without the inclusion of open biomass burning, but including biofuels). It is not clear if the suite of sources included in the two studies is the same. We estimate the distribution of Asian CO₂ emissions across sectors as: 27% residential (including biofuels), 23% industry, 23% power generation, 12% transportation, and 12% biomass burning. This contrasts with the larger proportion of transportation and smaller share of residential emissions in the West.

For China, we estimate CO₂ emissions to be 3820 Tg. This is the same as the value we reported in a separate study of recent trends in China's CO₂ emissions, allowing for the inclusion of carbon uptake by soils and growing vegetation in that study [Streets *et al.*, 2001b]. Other available estimates of China's CO₂ emissions [e.g., Tsinghua University and others, 1999; Wu and Wei, 1997] should be compared with caution, because they are usually for an earlier time period like 1990 and omit biofuel combustion and biomass burning. For India, we estimate 1890 Tg. Garg *et al.* [2001a] report 778 Tg CO₂ in 1995, but apparently without the inclusion of emissions from biofuel use or biomass burning. As with continent-wide emissions, there is little reliable data for individual countries, and the data that do exist are inconsistent with regard to inclusion of various types of sources and sinks.

3.1.4. Carbon monoxide. Carbon monoxide emissions in Asia are not known with any great confidence, because emission factors are highly dependent on the efficiency of the combustion process and the operation and maintenance of the equipment—none of which have ever been surveyed in a systematic manner. Throughout

most of developing Asia, equipment performance is poor and CO emissions are high. We estimate that Asian emissions of CO were 279 Tg in 2000, of which 67 Tg, or 24%, comes from open biomass burning. The energy component is largely due to residential biofuel combustion (96 Tg or 34%) and transportation sources (78 Tg or 28%). *Olivier et al.* [1996] report 290 Tg for 1990, but this value is believed to be high due to over-estimation of emissions from the burning of agricultural waste (see EDGAR emission inventory web site for a discussion of the differences between EDGAR Versions 2 and 3 at <http://arch.rivm.nl/env/int/coredata/edgar/index.html>.)

Our estimate for China is 116 Tg. This value is high because vehicles in the less-developed interior of China are relatively high emitters of CO [*Fu et al.*, 2001] and because emission factors for small coal combustors are 2-3 times higher than comparable sources in the West [*US EPA*, 2002]. There is a pressing need to undertake a program of field testing of sources in the region, particularly transportation and industrial, so that emissions of CO and other products of incomplete combustion can be quantified more reliably. The only other estimates of CO emissions in China are 87.4 Tg for 1994-1995 by *Tonooka et al.* [2001] and our earlier estimate of 113 Tg for 1995 [*Streets and Waldhoff*, 2000]. We believe that CO emissions in China are relatively stable today, as vehicles, industrial processes, and household energy sources with improved combustion efficiencies gradually replace outdated technology. We estimate CO emissions in India to be 63.3 Tg. Emissions in Southeast Asia are 65.7 Tg, of which almost 50% originates from biomass burning practices, especially slash-and-burn agriculture and deforestation.

The CO estimates for India are very sensitive to the assumed emission factors for biofuel combustion in small stoves. As discussed earlier, *Zhang et al.* [1999] and

Venkataraman and Rao [2001] agree that emission factors, especially for the burning of dung cake, are very high, but the magnitude is highly dependent on stove design and combustion conditions, both of which are poorly known for vast areas of developing South Asia. Emissions from Indian vehicles are also thought to be high, especially for two-wheelers and two-stroke engines in general [*Das et al.*, 2001]. We await publication of other estimates of CO emissions in India. *Bhattacharya et al.* [2000] have estimated the emissions of several species from biofuel use in Asian countries, and the results for CO emissions are informative, even though their results are characteristic of the early 1990s. The two sets of results are in good agreement for a number of countries, e.g., (our values first) China (36 Tg vs. 31 Tg), Pakistan (4.1 Tg vs. 2.8 Tg), Philippines (1.2 Tg vs. 1.4 Tg), and Sri Lanka (610 Gg vs. 740 Gg). However for India, our value is considerably higher (29 Tg vs. 9.9 Tg), as it is for Vietnam (4.3 Tg vs. 1.9 Tg). The differences are partly due to the use of different emission factors and partly due to differences in estimates of the amounts of biofuel used, with the countries that disagree the most having the greatest differences in biofuel usage estimates. CO emissions are further discussed in Section 3.6.

3.1.5. Methane. Estimation of CH₄ emissions in Asia requires consideration of many varied source types not considered for other species. In this work we estimate emissions from rice cultivation, animals (both manure and enteric fermentation), landfills, wastewater treatment, coal mining and combustion, oil and gas extraction and combustion, biofuel combustion, and biomass burning. Emissions from wetlands are considered a natural source and not included in this anthropogenic inventory. Our estimate of CH₄ emissions for all of Asia in 2000 is 107 Tg. The IPCC estimate is 125 Tg

[*Nakicenovic et al.*, 2000], while the EDGAR inventory reports 118 Tg for 1990 [*Olivier et al.*, 1996]. China is the largest emitter in our inventory at 38.4 Tg followed closely by India at 32.9 Tg. This estimate for China is higher than the value of 33.3 Tg that we reported in *Streets et al.* [2001b] due to the inclusion of several additional source types and our own estimation of biomass burning emissions rather than that of *Olivier et al.* [1996]. The estimate from the China Country Study is 32.4 Tg for 1990 [*Tsinghua University and others*, 1999].

We find that emissions from manure and enteric fermentation in animals represent the largest source of CH₄ emissions in Asia at 36.2 Tg (34%), followed by rice cultivation (24.2 Tg, 23%), wastewater treatment (14.0 Tg, 13%), landfills (8.9 Tg, 8.3%), biofuel combustion (8.6 Tg, 8.1%), coal mining and combustion (8.4 Tg, 7.9%), oil and gas extraction and use (3.4 Tg, 3.1%), and biomass burning (3.1 Tg, 2.9%). The distribution of CH₄ emissions among source types varies considerably from country to country. In China, for example, rice cultivation is the largest source type (26% of national total), while in India animal emissions dominate (54%), and in Japan and the Republic of Korea landfills are the largest source type (31% and 39%, respectively). These balances of source types are continually shifting as national priorities change. For example, oil and gas extraction and use is rapidly increasing in China; municipal waste incineration is replacing landfills in developed countries with limited land area, like Japan, Singapore, and Taiwan, China; and biomass burning is now declining in Southeast Asian countries.

It is interesting to compare our CH₄ values for China with other data sources. We estimate emissions of 9.8 Tg from rice cultivation. This is significantly lower than a recent estimate by *Verburg and Denier van der Gon* [2001] of 20.2 Tg for 1991. Our

method is based on the detailed analysis by *Tsinghua University and others* [1999], which yielded a value of 11.2 Tg. Even Verburg and Denier van der Gon acknowledge that “most recent estimates are ... between 9 and 15 Tg,” though they claim that rice harvested area is under-reported in Chinese statistics. Other poorly known factors like the degree of organic amendment, irrigation, and flooding introduce considerable uncertainty into the estimate (see also *Lin et al.* [1997]). For CH₄ emissions from enteric fermentation and manure management in China, our value of 8.9 Tg for 2000 is in good agreement with the value of 8.8 Tg for 1991 from *Verburg and Denier van der Gon* [2001]. Our estimate for CH₄ emissions in India is 32.9 Tg, dominated by emissions from animals, particularly cattle. This is considerably higher than the value of 18 Tg reported by *Garg et al.* [2001a] for 1995. Resolution of differences must await a detailed side-by-side comparison of emission factors and activity levels.

3.1.6. Non-methane volatile organic compounds. Emissions of non-methane volatile organic compounds (NMVOC) in Asia are subject to a high degree of uncertainty due to the great variety of sources, many of which are poorly known in terms of their operating characteristics and emission controls. Nor have many other attempts been made to estimate NMVOC emissions in Asia. We estimate anthropogenic emissions in all of Asia to be 52.2 Tg (measured as full molecular weights of the constituent compounds); this paper does not address biogenic emissions. Emissions are largest from the residential combustion of coal and biofuels (about 34%) and from transportation (27%). We believe that emissions of NMVOC are growing rapidly in Asia as countries extract and use petroleum products and natural gas in increasing amounts, expand the production of industrial organic chemicals, use paints and other surface coatings, and

develop high-technology industries that use solvents for cleaning. *Olivier et al.* [1996] estimated 1990 emissions of NMVOC in Asia to be 53.3 Tg; however, this value is believed to be high because of over-estimation of emissions from agricultural waste burning (see Section 3.1.3). A global inventory by *Piccot et al.* [1992] has a value of 20.4 Tg for a period characteristic of Asia in the mid-1980s.

Our NMVOC methodology for Asia is fully described in *Klimont et al.* [2002], which addressed China only. The year-2000 value for China in that report is 15.6 Tg. The China estimate presented in this analysis is 17.4 Tg, the difference being due to the addition of emissions from savanna and forest burning. To the best of our knowledge there are no estimates of NMVOC emissions in China made by Chinese researchers. *Tonooka et al.* [2001] calculated China's NMVOC emissions in 1994/1995 to be 13.9 Tg, without open biomass burning, which is quite consistent with this work. For all of East Asia, we estimate 21.7 Tg for 2000, compared with 17.1 Tg for 1995 by *Klimont et al.* [2001] and 17.7 Tg for 1994/1995 by *Tonooka et al.* [2001], both without full biomass burning emissions. So, again, agreement is reasonable. Outside East Asia there are no NMVOC emission estimates against which to compare, so we simply state that our estimate for all of Southeast Asia is 16.8 Tg and for India 10.8 Tg, both of which are quite uncertain due to lack of detailed information about industrial activities. Section 3.3 discusses the speciation of total NMVOC emissions into its chemical components.

3.1.7. Black carbon. Submicron black carbon emissions in Asia are estimated to be 2.54 Tg, with a high degree of uncertainty, as discussed in Section 3.5. The approach taken and emission factors chosen are consistent with *Streets et al.* [2001c] for China. The residential sector—consisting of the combustion of coal, kerosene, and

biofuels in stoves, cookers, and heaters—generates the majority (64%) of these emissions. The other large source is biomass burning (18%), especially in Southeast Asia. Because BC emission factors have been revised significantly since the earlier work of *Cooke et al.* [1999], as discussed in *Streets et al.* [2001c], their estimate of 2.04 Tg of submicron BC in Asia for 1984 without biofuels or biomass burning is not directly comparable with our work.

We estimate 1.05 Tg for China in 2000, significantly less than the value of 1.34 Tg we reported for 1995 [*Streets et al.*, 2001c]. This real reduction we attribute to a combination of several factors: economic downturn in the late 1990s, improved energy efficiency, the conversion of many residential users from raw coal to coal briquettes and cleaner fuels like LPG and electricity; and a concerted effort to clean up the air in China's more polluted cities through the closure of small coal-burning factories. As reported in *Streets et al.* [2001c], BC emissions in China are dominated by the residential sector (781 Gg, 74%), due to the continued direct combustion of coal and biofuels throughout much of China. Though emissions from individual diesel vehicles are high in China and the use of vehicles of all kinds grows rapidly, the transport sector is still a small part of the Chinese economy and contributes only 6% to national BC emissions. The value of *Cooke et al.* [1999] for China is 1.15 Tg (but see discussion above).

We estimate BC emissions in India to be 600 Gg, though this is even more uncertain than the estimate for China because of doubts about the emission factors of Indian vehicles, the degree of fuel adulteration in Indian vehicles, and emission factors for dung-burning cook-stoves. *Reddy and Venkataraman* [2002a,b] estimate Indian BC emissions to be 380 Gg for 1998-99, using a similar approach and emission factors to

ours, though the belief has been expressed in the literature that BC emissions could be considerably higher [*Dickerson et al.*, 2002]—possibly as high as 2-3 Tg—based on atmospheric measurements during INDOEX. While the source strength of BC is indeed uncertain, comparison between models and ambient measurements is further confounded by inconsistencies between different BC measurement techniques.

3.1.8. Organic carbon. Submicron organic carbon emissions in Asia are estimated to be 10.4 Tg C, with a high degree of uncertainty similar to BC. As for BC, residential fuel combustion represents the largest share of the Asian total (6.7 Tg, 65%), but for OC biomass burning is also large (3.3 Tg, 32%). All other sources of OC are relatively small. *Cooke et al.* [1999] report OC emissions of 2.44 Tg submicron OC for Asia in 1984, but the same caveats apply as for BC noted above; the values of *Cooke et al.* [1999] are 2-3 times lower than values obtained by *Lioussé et al.* [1996], which included biofuel combustion and open biomass burning.

For China we obtain an OC emission estimate of 3.38 Tg C, 2.57 Tg (76%) from residential fuel combustion and 0.73 Tg (22%) from biomass burning. These two source types are even more dominant for China. The value of *Cooke et al.* [1999] for China is 1.54 Tg. Our estimate for India is 2.84 Tg. This can be compared with 1.25 Tg for 1998-1999 from *Reddy and Venkataraman* [2002a,b]. Differences between emission estimates for both BC and OC highlight the need for more source testing and inter-comparison of measurement methods for primary aerosols.

3.1.9. Ammonia. The major sources of ammonia are not energy-related, so they require a greater emphasis on agricultural activities. The large quantities of fertilizer, specifically urea and ammonium bicarbonate (ABC), used in China make this activity a

big source of NH_3 emissions. We estimate that NH_3 emissions in Asia in 2000 were 27.5 Tg. *Zhou and Wang* [1994] estimated that annual Asian NH_3 emissions for the period 1989/1991 were 24.6 Tg, while the global inventory of *Olivier et al.* [1998] gives 22.5 Tg for 1990. Growth in agricultural production and fertilizer application in the decade of the 1990s can account for the increase. Similar growth rates for East Asian NH_3 emissions are reported by *Klimont et al.* [2001]. Our shares of Asian NH_3 emissions are: fertilizer application 45%, animals 38%, miscellaneous sources (humans, pets, waste disposal, fertilizer manufacture, etc.) 7.7%, biofuel combustion 6.1%, and biomass burning 3.3%.

For China, we estimate NH_3 emissions in 2000 were 13.6 Tg, of which 50% comes from fertilizer application and fully 88% from the agriculture sector as a whole. This is in good agreement with *Zhou and Wang* [1994], though their China estimate of 13.6 Tg in 1989/1991 is larger than would be consistent with our approach. This can be largely explained by a discrepancy in estimates of fuel combustion; *Zhou and Wang* [1994] estimated significantly larger emissions from this source. *Bouwman et al.* [1997] agree with us that fuel combustion is a minor source of NH_3 . Our estimate for NH_3 emissions from agriculture in Japan, 266 Gg, agrees better with the estimate for 1990 of *Murano et al.* [1995] of 200 Gg than the higher value for Japan of *Zhou and Wang* [1994] of 346 Gg for 1989/1991. For India our estimate is 7.4 Tg NH_3 .

It needs to be stressed that estimates of NH_3 emissions carry a significant uncertainty linked to the use of European-based emission factors (especially for livestock). This applies to studies performed by both European and Asian researchers. Practices in the Asian livestock industry are different from those in Europe, but there is insufficient information to adjust European emission factors for the important variables

since measurements in Asia are lacking. This observation can generally be applied to a number of sources and species in this inventory (particularly NMVOC) though not all.

3.1.10. Inter-comparison of emissions. Table 6 summarizes the emissions of each species in each Asian country and shows that China dominates emissions of most species. China contributes the following percentages of total Asian emissions of each species: 59% SO₂, 42% NO_x, 39% CO₂, 42% CO, 36% CH₄, 33% NMVOC, 41% BC, 32% OC, and 49% NH₃. India also contributes significantly to the totals, as follows: 16% SO₂, 17% NO_x, 19% CO₂, 23% CO, 31% CH₄, 21% NMVOC, 24% BC, 27% OC, and 27% NH₃. Other countries contribute much smaller individual shares, though Japan becomes important for CO₂ (12%).

Figure 3 shows the shares of Asian emissions among different source regions. Southeast Asia contributes significantly to emissions of CO, NMVOC, and OC, due to the strong component of biomass burning. South Asia, including India, is a major contributor to CH₄ and NH₃ emissions, because of the agrarian component, especially livestock and rice production. Other East Asian countries, being at a higher level of economic development and industrialization, contribute most to NO_x and CO₂ emissions.

Figure 4 is particularly informative in its depiction of the source types that contribute most to the emissions of each species. It can be seen that the traditional air pollution sources—industry, power generation, and transportation—indeed contribute most to the traditional air pollutant species that have been most studied in the West, SO₂ and NO_x. However, for carbonaceous species, there is much more involvement of other source types in Asia. Emissions of the primary aerosol species BC and OC, for example, are dominated by the residential and biomass burning sectors. And emissions of CH₄ and

NH₃ are dominated by agriculture. This means that much greater attention has to be paid to these source types in Asia than has been devoted to them in the West.

3.2. NMVOC Speciation

A detailed analysis of the chemical speciation of NMVOC emissions has been performed. As described in the methodology section, we have speciated the NMVOC emissions into 19 categories based on chemical reactivity and functional groups. This representation was performed for each geographical area in the region for each of 82 emitting source types [Klimont *et al.*, 2002]. The speciation profiles for each source type were drawn from four major information sources: the USEPA SPECIATE database [EPA, 2000], the CORINAIR inventory [EEA, 2000], *Smith et al.* [1992] for the special case of Asian biofuel stoves, and *Andreae and Merlet* [2001] for open biomass burning. For the first two information sources, we recognize the implicit assumption that the species profile for a given source type in Asia is the same as for the same source type in the West; this may or may not be a reasonable assumption, but with a complete lack of VOC emissions test data for most sources in the Asian setting we have no other choice.

Table 7 presents the distribution of aggregated chemical species groups by country. In summary, for all of Asia we estimate that 27% of NMVOC emissions are alkanes, 22% alkenes, 5% ethyne, 17% aromatics, 9% aldehydes and ketones, and 20% halocarbons and other organic compounds. This kind of distribution is typical of all of Asia except the developed or industrializing parts of East Asia (such as Japan; the Republic of Korea; and Taiwan, China). In this region, the share of alkanes is higher (about 35%) and the share of alkenes is much lower (about 9%). This is indicative of

greater use of petroleum products, particularly for transportation, and far less vegetation burning. *Piccot et al.* [1992] calculated the following shares for Asian NMVOC in the mid-1980s: alkanes 46%, alkenes 30%, and aromatics 22% (others not matching our categories). This higher share of hydrocarbons might be due to relatively greater combustion activities (fossil fuels and biofuels) in the mid-1980s and less use of industrial chemicals and solvents [*Klimont et al.*, 2002]. Figure 5 shows the species profiles of emissions from four different countries. China and India have remarkably similar profiles, reflecting similarities in the state of development and resource use. Japan, on the other hand, shows a much larger fraction of alkanes, particularly butanes and $>C_5$ alkanes, indicative of a petroleum-rich, developed economy with heavy transportation needs; emissions of alkenes, aldehydes, and ketones are much lower. The case of Japan can be contrasted with that of impoverished Laos, where alkane emissions are very low and emissions of alkenes, aldehydes, and ketones are high.

Figure 6 shows the major sources of some important compounds for East Asia. Biofuel use dominates the emission profiles of ethane (45%), ethene (50%), and ethyne (45%). Emissions of butanes are primarily from transportation (58%). Formaldehyde emissions (56%) are mainly from biomass burning, and toluene emissions (45%) are mainly from industry. Such data are important for identifying the major source regions that contribute to observed concentrations of particular species.

3.3. Seasonality of Emissions

There is undoubtedly seasonal dependence of emissions, though this is not easy to determine. Generally, we can expect that the space heating component of residential

energy use will depend on outdoor temperature, though the cooking component will not. For the purposes of illustrating the seasonal variation of emissions, we have examined the potential monthly variation of residential energy use in China, while assuming that other components remain constant. (Of course, this is a big assumption, because we can expect that power generation will also have some seasonal variation in China, as well as industrial energy use; however, there is no information available to estimate these changes—China does not report provincial fuel use by sector and month.) We developed monthly mean temperatures for each province of China from *Lin* [1995]. We then assumed the following dependence of stove operation as a function of provincial monthly mean temperature: $< 0^{\circ}\text{C}$, 16 hr/d; $0\text{--}5^{\circ}\text{C}$, 12 hr/d; $5\text{--}10^{\circ}\text{C}$, 6 hr/d; $>10^{\circ}\text{C}$, 3 hr/d. The last value in this list is meant to approximate cooking-only stove usage, based on the work of *Ravindranath and Ramakrishna* [1997]. Annual emissions at provincial level were then shared out by month according to this residential stove usage scheme.

Figure 7 shows the results for the primarily combustion-related species SO_2 , NO_x , CO, NMVOC, and BC. The strongest seasonal dependence is observed for BC and CO, because residential fuel use constitutes the largest contributor to total emissions for these species. We find a peak in January, with a significant shoulder in the period February–March. The ratio of monthly emissions in January to emissions in June, the minimum, is approximately 1.9. We find that SO_2 and NO_x emissions have very similar seasonal dependence, with a ratio of 1.2 between maxima and minima. Emissions during the TRACE-P and ACE-Asia months of March and April are almost exactly at the monthly average fraction of 0.083 of annual emissions, so detailed seasonal analysis is not necessary for these campaigns; however we would expect that observations should be

higher in March than in April by perhaps 10%. Figure 7 also shows the latitudinal dependence of monthly CO emissions for northern China (defined as provinces north of the Huang He or Yellow River), southern China (provinces south of the Chang Jiang or Yangtze River), and central China (provinces in between these two rivers). Perhaps surprisingly, the greatest seasonal dependence is observed for the central region, because of the sharper contrast between summer and winter. In the other two regions there are longer cold and warm seasons which flatten out the seasonality of emissions.

Seasonality of CH₄ and NH₃ emissions in China has also been determined. In these cases we examined the temperature dependence of emissions of CH₄ from animal waste and rice cultivation and of NH₃ from animal waste and fertilizer application. For rice cultivation we considered the planting cycles of various rice crops by province, and for fertilizer application we considered the application of fertilizer prior to the planting of wheat, corn, and rice by province. The seasonality of CH₄ emissions from domestic fuel combustion was also incorporated. The results are shown in Figure 8. The profile generally is the opposite of the profiles of combustion-related species (Figure 7). NH₃ emissions peak in June, and CH₄ emissions peak in August. The greatest seasonal variation in NH₃ emissions (Figure 8) is observed in northern China, but there is no great difference by latitude.

3.4. Gridded Emissions

Spatial allocation (gridding) is a process to transform large and irregularly shaped emission data to relatively small and uniform (usually rectangular) data. Since air pollution is not a spatially uniform problem, spatial allocation with different resolutions

is important for different levels of model application and analysis, i.e., at global, regional, urban, and local scales.

For this inventory, we developed a multi-resolution, spatial allocation technique using Geographical Information System (GIS) and Remote Sensing (RS) technology to support the various scales of research needs for TRACE-P and ACE-Asia. Basically, we use a “top down” approach to break regional- or country-based emission data down to their various gridded forms. This method is adapted from previous research, but further advanced in terms of calculation efficiency and flexibility (*Woo et al.*, submitted manuscript, 2002). Since the “top” emissions data are different for each administrative region and sector, the allocation factors are separately developed for each category so that emissions can be faithfully allocated “down” to each grid cell. To generate the allocation factors, various types of GIS/RS datasets are used: (1) for common use, administrative boundaries are extracted from the RAINS-Asia model [*Foell et al.*, 1995; *Shah et al.*, 2000] and the Digital Chart of the World (DCW) [*Defense Mapping Agency*, 1989]; (2) for large point sources, exact geographical coordinates are taken from the RAINS-Asia model and the GEIA inventory [*Graedel et al.*, 1993]; (3) for line sources, road networks and ship lanes are extracted from the Digital Chart of the World and previous research by *Streets et al.* [2000b]; (4) for other area sources, high-resolution (30 sec \times 30 sec) population and land-cover data are extracted from the LandScan Global Population database developed by Oak Ridge National Laboratory [*ORNL*, 2001].

The spatial allocation of large point sources is a two-step process. First, the regional emission total is subdivided into the sum of large point source emissions and small area source emissions in the appropriate sector. Second, the sum of large point

source emissions is allocated to each facility on the basis of plant size or plant production. In the present scheme, large point sources are primarily power plants, except that large iron and steel plants are included for CO emissions. Line and area source emissions allocation is conducted in one simple step, by multiplying pre-generated allocation factors by regional or national emission totals. The allocation factors for line and area sources are created by combination and transformation of geo-registered information, such as road network type (e.g., connector, dual-lane highway, primary or secondary roads, and tracks, trails or footpaths), ship-lane information (e.g., major/minor ship lanes and harbors), population distribution (total, urban, and rural), land-cover classes (urban/suburban, crop fields, forest, savanna, grassland, and wetlands). For example, CO emissions from biofuel combustion are allocated by rural population distribution, since most biofuels are consumed in rural households. After the spatial allocation of all emissions by chemical species and sectors is completed, sectoral aggregation is conducted to sum emissions up to two emission types, point and area, for the practical purpose of reducing the number of unnecessary files. Special emission allocation schemes can be developed for special research needs, as necessary.

Figure 9 shows the area source emission distribution of four of the gaseous species: SO₂, NO_x, NMVOC, and NH₃, at 30 min × 30 min resolution. These figures are presented at the same scale, so comparative observations can be made. A number of features stand out: the source strengths of NH₃ in the agricultural areas of central China and northern India, the relatively strong signature of NO_x from transportation sources in Japan, significant emissions of NMVOC from biomass burning in Southeast Asia, and large SO₂ emissions in the Sichuan Basin and industrialized eastern China. In Figure 10,

BC and OC emissions are compared, also at $30 \text{ min} \times 30 \text{ min}$ resolution and at the same scale. This figure shows how much stronger the OC signal is than BC throughout the region, as well as the broad distribution of both species across rural areas of China, where domestic coal and biofuel combustion are prevalent. Figure 11 presents the distributions of four gaseous organic chemical species: total alkanes (except CH_4), ethene, toluene, and formaldehyde. Note that the two upper maps are at the same scale, but the two lower maps are at a different scale. These graphs conform to Figure 6, in revealing the dominant source associations of these species: alkanes and ethene with biofuel combustion, toluene with industry, and formaldehyde with biomass burning. These maps are specifically for the month of January, and therefore they emphasize emissions from domestic heating in China and biomass burning in Southeast Asia.

In terms of spatial resolution, the initial allocation procedures are conducted at the highest possible resolution ($30 \text{ sec} \times 30 \text{ sec}$ —about $1 \text{ km} \times 1 \text{ km}$) then aggregated to lower resolution to meet the needs of modeling and analysis. In theory, any resolution is possible that is lower than or equal to $30 \text{ sec} \times 30 \text{ sec}$. Figure 12 shows SO_2 and CO emission distributions at the highest resolution in the Pearl River Delta, as an example. In this figure, the effects of regional and sectoral differences can be clearly seen for the two different chemical species. This makes focused studies of large metropolitan air-sheds feasible with this inventory.

3.5. Uncertainty in Emission Estimates

We acknowledge that for some types of sources in some countries very little is known about actual activity levels and emission factors, and our choices for this

inventory in such cases rely heavily on inferences of activity levels from quite limited and uncertain statistical information and extrapolations of emission factors from western or other Asian source types. Several factors influence the estimate of emissions, including emission factors, activity levels, sulfur content of fuel, and removal efficiency of abatement technology. We estimate the error for each emitting sub-sector by combining the coefficients of variation (CV, or the standard deviation divided by the mean) of the contributing factors. We then combine these uncertainties to estimate the uncertainty of emission estimates for each country. We tried wherever possible to use harmonized data across different source types and regions, even though the reporting practices of countries are not always consistent and available.

Since there is no way to judge the accuracy of activity estimates, expert judgment was used, classified according to a ten-point scale of CV, which varied for each group of provinces or countries based on the level of economic development and perceived statistical quality. For example, coal consumption estimates for Japan and Hong Kong are believed to be highly reliable and are assigned the lowest value of $\pm 5\%$; at the other extreme, the quantity of grassland burned in Myanmar is extremely difficult to estimate reliably and assigned the highest value of $\pm 1000\%$. For the CV of emission factors, the available literature sources that reported emission factors based on laboratory tests usually included standard deviations that could be used. In some cases no uncertainty estimate was provided with the average value, so expert judgment was again used. Whenever emission factors were taken from the USEPA's AP-42 compilation [USEPA, 2002], the letter-grade emission factor ratings of A~E were transferred to CV of $\pm 5\% \sim 500\%$ in our estimations. We have assumed that the underlying emission factor

measurements are normally distributed [Suutari *et al.*, 2001], but a case can be made for a lognormal distribution of combustion emission measurements (T.C. Bond *et al.*, manuscript in preparation, 2002).

Combining uncertainties requires assumptions about the dependence or independence of parameters. To combine multiplicative, independent random variables such as activities and emission factors, we used Goodman's formula for the product of variables. The relative 95% confidence intervals for emissions are calculated as 1.96 times CV. However, combining uncertainties across emitting sectors or provinces within a country is more problematic. When uncertainties are independent they can be combined in quadrature; this can be done for different sectors such as industry, residential, agriculture, biomass burning, etc. However, since the same emission factors are used to calculate the emissions from two provinces, for example, these uncertainties are not independent and should not be added in quadrature. The same is true of two different sources for which an identical emission factor is used. For each sector, then, uncertainties are added linearly when the same emission factor is used, and uncertainties in emissions from different provinces are also added linearly. These aggregated uncertainties are then independent of each other and are added in quadrature.

Table 8 and Figure 13 show the results for each species for seven prototypical Asian regions. Our general findings are that emissions are known least well in India, the rest of South Asia, and Southeast Asia. This has much to do with the large contributions of biomass burning and uncertainties about liquid fuel consumption in vehicles. Emissions are best known in Japan and the other East Asian countries. The overall uncertainty in emissions for all of Asia is as follows, ranked in increasing order of

uncertainty and measured as 95% confidence intervals: $\pm 16\%$ (SO_2), $\pm 31\%$ (CO_2), $\pm 37\%$ (NO_x), $\pm 65\%$ (CH_4), $\pm 72\%$ (NH_3), $\pm 130\%$ (NMVOC), $\pm 185\%$ (CO), $\pm 360\%$ (BC), and $\pm 450\%$ (OC). So, for example, we are 95% confident that Asian emissions of SO_2 are within $\pm 16\%$ of the stated value. The large uncertainty values for the primary aerosol species BC and OC arise from the high variability of measured emissions among tests under varying combustion conditions. The values for Asian emission uncertainties are similar to recent estimates made for some European countries [Suutari *et al.*, 2001]. In that study, the following ranges of 95% confidence intervals were obtained for emissions in European countries: ± 6 -23% for SO_2 ; ± 9 -26% for NO_x ; and ± 9 -23% for NH_3 . Because of the more meticulous record-keeping and source testing in Europe, we would expect lower uncertainties than in Asia.

3.6. Discussion

This paper presents an inventory of air pollutant emissions from Asian sources. It uses available information on source activity rates and emission factors and is compared with results from other inventories where they are available. All of this information seems to be internally consistent. However, we would really like to know how robust the inventory is in representing actual source strengths in the real world. The inventory implicitly consists of thousands, in some cases millions, of individual pollution sources—most of which have never been tested—scattered across a very wide area. Is there any evidence from observations and computer simulations that can substantiate the inventory?

One verification method that is being pursued in TRACE-P, ACE-Asia, and similar experiments, is to input the emissions data into atmospheric models and compare

the modeled concentration values with field observations at the same times and places. Consistency may increase confidence in the reasonableness of the emission values; inconsistency may cast doubt on the inventory. However, in the latter case one should not automatically assign problems to the emission estimates, because the models have their own sets of assumptions and shortcomings and the measurements themselves are not always reliable or directly comparable with model predictions.

Initial results from the TRACE-P and ACE-Asia experiments are encouraging. Table 9 presents a comparison between observed mean concentrations of ten selected species and concentrations simulated with the STEM model [*Carmichael et al. (a)*, this issue] using this emission inventory. In Table 9, values are included for TRACE-P DC-8 flights #6 through #17. Values are limited to altitudes below 1 km, because the concentrations at these altitudes often reflect sampling directly downwind of large sources (in the cases of flights in the Yellow Sea) and are not as influenced by complicating factors such as vertical transport by convection. The general agreement in Table 9 between observed and modeled values for a range of organic and inorganic species is reassuring. A more detailed table with more species and more comparison criteria can be found in *Carmichael et al. (a)* [this issue]. Similarly, *Zhang et al.* [this issue] found that the CMAQ model using this inventory reproduces well the latitudinal and vertical distribution of CO, O₃, and SO₄²⁻ in the Asian outflow.

One issue concerns the SO₂ and SO₄²⁻ observations. The SO₂ concentrations measured in the Yellow Sea were very high during TRACE-P, and significant increases were noted between PEM-West B (1994) and TRACE-P, even though SO₂ emissions have declined during the intervening time period in East Asia [*Streets et al.*, 2000a]. This

is not believed to be a problem with the anthropogenic emissions, however, but more likely due to different precipitation regimes affecting the conversion of SO_2 to SO_4^{2-} in the two campaigns or the influence of the Miyakejima volcano during TRACE-P. To further assist researchers in interpreting measurements in Asian field campaigns separated by a number of years, Table 10 is presented. This table shows trends for seven major species in the 1990-2000 period, with emphasis on differences between 1994 (PEM-West B) and 2000. A brief explanation of the reasons for the trends is included. Emissions of two species (SO_2 and BC) have declined in the latter half of the 1990s, two have changed little (CO_2 and CH_4), and three have increased significantly (NO_x , NMVOC, and NH_3). Though we do not have trend information for CO, we believe that CO emissions have been stable in recent years (Streets and Waldhoff, 2000).

Some initial concern was expressed that BC emissions were too low in comparison with observations during TRACE-P and ACE-Asia. However, further reassessment of the observational data seems to yield better agreement. BC concentrations modeled using the CFORS model and this inventory have been compared with observed BC concentrations measured during the ACE-Asia period at five Japanese island sites: Rishiri, Sado, Hachijo, Chichijima, and Amami-Oshima (I. Uno *et al.*, manuscript in preparation, 2002). Very good agreement is observed for both absolute concentration levels and time variation of the concentrations. Occasionally at Sado there is an under-prediction when the prevailing wind is from the main Japan islands. This suggests that local Japanese sources are partly responsible, and the authors conjecture that the burning of agricultural residues in the field prior to rice planting may be the cause. Also Clarke *et al.* [this issue] have shown good agreement between measured

absorption on TRACE-P flights #8 through #19 and modeled BC concentrations using this inventory. Averages of observations grouped in $0.1 \mu\text{g m}^{-3}$ bins fit well within the bounds of $6\text{-}10 \text{ m}^2 \text{ g}^{-1}$ BC.

The major pollutant of contention is CO. Table 9 showed that mean CO concentrations simulated with the STEM model are in good agreement with the observational data set. Figure 14 elaborates on this simulation by showing the STEM model/observation inter-comparison of all 5-minute values for DC-8 flights #6 through #17 and P-3 flights #8 through #19. The results show that approximately 80% of the predicted values fall within $\pm 30\%$ of the observed values. However, there is a slight bias towards under-prediction, especially at very high CO concentrations. Approximately 7% of values are under-predicted by more than 30% and 1.4% of values are under-predicted by more than 50%. In particular, it seems that some of the very high concentrations observed during flights close to the coast of China are not well reproduced. This poses the question of whether emissions of CO are under-estimated in the inventory.

Analysis by global modelers also suggests that CO estimates for China are low. The Asian emission inventory for CO described here was used in the GEOS-CHEM model to simulate observations of CO during the TRACE-P campaign [*Palmer et al.*, this issue]. A comparison of the modeled and observed distribution of CO showed that the model had a significant negative bias, especially in the boundary layer. An inverse model analysis of these data suggested that an increase in modeled anthropogenic emissions (fossil fuel and biofuel) over China largely eliminates the model bias.

It is possible that model resolution could explain the differences between the regional and global models. For example, *de Laat et al.* [2001] noted that during the

INDOEX program their high-resolution model ($1.9^{\circ} \times 1.9^{\circ}$) was much better able than their low-resolution model ($3.75^{\circ} \times 3.74^{\circ}$) to reproduce measured CO mixing ratios. They further state that the under-estimation of CO in the model for air masses from the Bay of Bengal is resolution-related, because discrepancies disappear at the higher resolution. This could explain some of the differences between the global and regional CO models. However, one might still expect any disagreement to manifest itself as greater scatter rather than bias in the inter-comparison. There are also reasons other than model resolution why the global model might not reflect local-scale phenomena satisfactorily, including incorrect levels of incoming pollution in the global model that are better represented in the boundary conditions of the regional models.

Additional evidence is provided by ground-station measurements in China. *Wang et al.* [2001, 2002] have measured concentrations of CO, NO_x, and SO₂ at an essentially rural site in eastern China—Linan in Zhejiang Province—for more than one year. They have also compared their observations with emission ratios from our inventory. The very high levels of CO concentrations led the authors to speculate that biofuel combustion was under-estimated in our inventory. While it is true that biofuel consumption is not well known, it is tracked by Chinese energy experts and presented in energy statistics [SSB, 1998]. We have examined the question of biofuel usage for the province in which Linan is located. The amount of CO released from biofuel combustion in Zhejiang Province according to our inventory is 1370 Gg, representing 49% of total CO emissions in the province. This estimate is obtained using the high emission factor estimate of 77.5 g kg^{-1} for biofuel combustion [*Zhang et al.*, 1999], combined with Chinese energy statistics. As a check, we performed our own analysis of the amount of biofuel used in Zhejiang

Province under a variety of assumptions about the number of biofuel stoves in the province, daily stove usage for cooking, and amount of biofuel consumed per day. Using the same emission factor, we obtained a range of emissions from biofuel combustion of 1110-1900 Gg CO. This analysis suggests that biofuel use is probably captured reasonably well in Chinese statistics.

Recent analysis of the Linan data has compared the measurements of *Wang et al.* [2001, 2002] with modeled concentrations using the STEM model and this inventory [*Carmichael et al. (b)*, this issue]. Significant under-estimation of not only CO but also SO₂ and NO_y is found. This would suggest that the under-prediction is more likely to reflect an under-estimation of coal use, rather than biofuel use. In addition, back-trajectory analysis of the observed CO concentrations during TRACE-P [*Carmichael et al. (b)*, this issue] seems to identify the central heartland of China as being the source of many of the air masses in which under-prediction is manifested. In combination, this evidence suggests an under-estimation of domestic coal use in rural China.

This is an intriguing question. Official Chinese energy statistics—on which our emission estimates are based—show a dramatic decrease in household use of coal in the late 1990s. From 135 million tons (mt) in 1995, it dropped to 122 mt in 1997 [*Sinton and Fridley*, 2000], to 89 mt in 1998 [*NBS*, 2000], and to 75 mt in 2000 (Jiang Kejun, Energy Research Institute of China, personal communication, 2001). This decline was taken up mainly by an increase in the use of LPG and electricity. While the trend is verifiably true in the developed coastal areas of China, it is difficult to believe that it could be true in the central heartland of China, where rural population continues to rise and the penetration of clean residential fuels is slow (J. Sinton, Lawrence Berkeley National Laboratory,

personal communication, 2002). In addition, though coal production has declined in this region according to official statistics, illicit mining is thought to be widespread, and much of that coal could find its way into rural households. So there certainly is the possibility for rural coal use to be under-reported in this region. The same argument can be applied to the use of coal to fuel small industrial operations, such as kilns and ovens. In addition, the higher temperatures of these applications can generate more NO_x, which seems to be indicated by analysis of the Linan data. It should be noted here that the problem may be restricted to this region of China where small sources dominate the profile of emissions. In other regions, emissions are larger and dominated by big industrial and power generation sources. This would explain why we do not always see discrepancies between models and observations—only when air masses originate in the rural heartland of China.

Another possibility is that there might be significant sources of emissions that are omitted from this inventory. Possibilities include open waste burning, coal-mine fires, and small coke ovens. We considered carefully whether open waste burning (of garbage) is significant in China. We believe that it is not. The amounts of garbage generated in China, especially rural China, are low compared to western countries; garbage tends to have higher moisture content than in the West and be more difficult to burn; and land disposal is a preferred option [*Wang and Nie*, 2001]. Observation around the Linan site revealed no major waste burning (T. Wang, Hong Kong Polytechnic University, personal communication, 2002). Though coal-mine fires have been touted as a major environmental catastrophe in China, Chinese and other western authorities believe that the amount of coal consumed in out-of-control burning is probably about 14 million tons annually, yielding 490 Gg CO or 0.4% of China's total CO emissions in this inventory

(A. Prakash, University of Alaska, personal communication, 2002, based on unpublished work by the Aerophotogrammetry and Remote Sensing Bureau of China). Also, many of the fires are in locations quite remote from Linan. Another possibility relates to the use of small coke ovens in eastern and central China, particularly in Shanxi Province, which are known to be high emitters of CO [*Polanske and McMichael*, 2002]. Unfortunately, we have no information on the number or emission rates of such facilities. However, this could be a way to increase CO emissions without increasing emissions of other species.

Is it possible that open biomass burning is under-estimated? We can likely rule out the extensive forest and agricultural burning that occurs in Southeast Asia, because these plumes are easily recognizable in the aircraft observations; also, they generally occur at high altitudes over eastern China and would not explain elevated ground-level CO concentrations at Linan. It is true that we know very little about farmers' practices as regards the burning of crop residues in the fields after harvest. Our calculations assume 17% of crop residues are burned in the field [*Hao and Liu*, 1994], somewhat less than the estimate of 25% by *Crutzen and Andreae* [1990], though to our knowledge neither value is based on surveys of agricultural practice. Actually, it is our understanding that crop burning in the fields is now banned in many (all?) parts of China but persists nonetheless. Visual observation around Linan has indeed revealed burning fields at some times of year (M. Bergin, Georgia Institute of Technology, personal communication, 2002); however, if this were the cause, one would expect a seasonality to the CO concentration profile that mirrors the growing and harvesting seasons (around Linan, say). The CO data of *Wang et al.* [2001], though, show no such seasonality—merely a small winter peak that is likely associated with a modest amount of additional home heating in Zhejiang and Jiangsu

Provinces and more northerly areas. Cross-sectional analysis of K^+ or CH_3Cl concentrations with other species or BC/OC ratios could help to elucidate whether biomass burning is the cause.

Finally, is it possible that the high CO is a product of unusual chemical reactions in the atmosphere? Perhaps the oxidation of biogenic hydrocarbons like isoprene emitted from the extensive southern forest and woodland areas of China is greater than anticipated? However, then a summer peak in CO concentrations might be expected, which is not observed. Alternatively, it has been noted by other TRACE-P researchers that peroxy and hydroxyl radicals over China are lower than expected, perhaps leading to less CO-destruction capability. More study of these possibilities is warranted.

We conclude with the following observations. This emission inventory for Asia is the first to treat many species in many countries with a consistent, detailed methodology for a recent year. Our methodology is internally consistent and seems to agree well with the few previous, reliable estimates that have been made. The inventory performs well for most species when input into atmospheric models and compared with observations. The possible exception to this is CO, for which some indicators suggest that our estimate is low. In some circumstances, other species like SO_2 and NO_x may be low as well. The most likely explanation is that Chinese statistics under-report coal use in residential and small industrial applications in the rural heartland of China. This is not a conclusive determination, however, and other possibilities are presented. Further analysis of the field campaigns in Asia may unravel this mystery. However, field testing of sources and surveys of source types and operating characteristics are the best ways to develop a better understanding of emissions in the region.

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Figure Captions

Figure 1. Definition of the inventory domain, its constituent countries, international shipping lanes, and sub-regions (vertical lines = China; vertical lines + dark gray shading = East Asia; light gray shading = Southeast Asia; horizontal lines = South Asia).

Figure 2. Schematic methodology for the development of Asian emission estimates.

Figure 3. Species emissions by Asian region and absolute values of emissions.

Figure 4. Species emissions by emitting sector.

Figure 5. Fractional emissions of NMVOC species in four selected countries.

Figure 6. Source contributions to emissions of six selected organic species in East Asia.

Figure 7. Seasonality of five primarily combustion-related species in China (upper) and CO emissions by latitude in China (lower).

Figure 8. Seasonality of two primarily noncombustion-related species in China (upper) and NH₃ emissions by latitude in China (lower).

Figure 9. Area source emission distributions at 30 min × 30 min resolution of selected gaseous species: SO₂ (upper left), NO_x (upper right), NMVOC (lower left), and NH₃ (lower right). The scale is the same for each species in units of Mg yr⁻¹ per grid cell.

Figure 10. Area source emission distributions at 30 min × 30 min resolution of primary aerosol species: BC (upper) and OC (lower). The scale is the same for each species in units of Mg yr⁻¹ per grid cell.

Figure 11. Area source emission distributions at 30 min × 30 min resolution of selected organic species: total alkanes (upper left), ethene (upper right), toluene (lower left), and formaldehyde (lower right). Scale is in units of moles sec⁻¹ per grid cell, different for the upper and lower pairs.

Figure 12. Illustration of sector-specific detail in the high-resolution (30 sec × 30 sec) inventory for the Pearl River Delta (at the lower right in each frame is Hong Kong; near the upper left is Guangzhou). Upper frame is SO₂; lower frame is CO.

Figure 13. Uncertainty (%) in pollutant emission estimates (±95% confidence intervals).

Figure 14. Comparison of modeled to observed CO mixing ratios for each five minutes along the flight paths for DC-8 flights #6 through #17 and P-3 flights #8 through #19.

Table 1. Fuel Use and Vegetation Burned in Asia in 2000

Country	Industry (PJ)			Domestic (PJ)			Transport (PJ)	Power (PJ)			Biomass Burned (Tg)		
	Coal	Oil	Other	Coal	Biofuel	Other	All fuels	Coal	Oil	Other	Savanna/ Grassland	Forest	Crop Residu
ina	5966	2523	2011	2417	7178	998	4246	9806	873	183	52	25	1
an	428	2490	1447	22	53	1506	3834	1783	2044	1757	0	1	
rea, Rep. of	232	1042	281	0	13	810	1175	752	373	230	0	0	
rea, DPR	651	12	21	0	294	24	57	199	0	0	0	1	
ngolia	32	14	22	0	22	59	9	51	4	0	23	9	
wan, China	239	585	186	0	6	136	485	466	316	113	0	0	
nei	0	12	97	0	2	7	19	0	0	25	0	0	
mbodia	21	6	8	0	80	3	25	12	3	0	8	5	
onesia	51	454	1059	0	1838	550	969	371	102	218	21	68	
os	0	9	0	0	34	0	15	0	0	0	5	19	
aysia	31	295	136	0	101	71	389	36	102	320	0	22	
anmar	0	14	87	0	348	12	52	0	2	39	2	56	
ippines	21	219	183	0	250	109	302	152	105	0	0	17	
gapore	0	238	138	0	0	3	94	0	200	70	0	0	
ailand	183	243	648	0	319	70	908	320	206	341	12	36	
etnam	70	82	42	19	850	26	101	63	21	23	12	15	
ngladesh	5	17	332	3	460	83	68	15	9	160	0	9	
utan	0	2	0	0	19	1	5	0	0	2	0	1	
ia	2085	893	1144	5	6740	841	2383	4597	133	350	9	37	
pal	5	16	4	1	259	13	11	0	2	0	0	5	
cistan	89	137	423	15	786	204	391	95	209	194	3	1	
Lanka	0	17	19	0	136	21	52	0	5	0	0	4	
ernational Shipping	0	0	0	0	0	0	726	0	0	0	0	0	
ia Total	10109	9320	8289	2482	19788	5548	16316	18718	4710	4024	147	330	2

Table 2. Non-Combustion Activities Included in the Inventory

Species	Activities Included					
CO ₂	Cement production					
CO	Steel production	Pig iron production				
CH ₄	Rice cultivation	Animal emissions	Landfills	Wastewater treatment	Coal mining	Leakage from oil and gas extraction and use
NMVOC ^a	Vehicle refueling and evaporation	Oil and gas extraction processes	Solvent and paint use	Chemical industry	Miscellaneous industry	Waste disposal
NH ₃	Cattle	Pigs	Other animals	Fertilizer application	Waste treatment and disposal	Nitrogen fertilizer manufacturing

^a 82 source categories were considered for NMVOC. See *Klimont et al.* [2002].

Table 3. Numbers of Motor Vehicles by Country in Asia in 2000 ($\times 10^3$)

Country	Cars		Buses		Trucks				Tractors	Motor-cycles
	LDGV (Gas)	LDDV (Diesel)	HDGV (Gas)	HDDV (Diesel)	LDGT (Gas)	LDDT (Diesel)	HDGV (Gas)	HDDV (Diesel)		
China	6205	773	368	217	9161	969	2075	2129	10124	13754
Japan	38976	15082	2	233	5089	4831	472	10019	144	13785
Korea, Rep. of	6926	2680	8	840	640	607	59	1259	161	3000
Korea, DPR	164	2	10	6	249	28	59	59	235	209
Mongolia	40	1	3	2	22	2	5	5	9	17
Taiwan, China	3764	6	0	23	177	168	16	349	169	11168
Brunei	166	0	0	2	4	6	0	4	3	5
Cambodia	68	0	0	1	5	7	0	4	1	568
Indonesia	2835	71	142	570	454	681	49	438	814	13635
Laos	11	0	0	1	2	3	0	2	20	356
Malaysia	3481	5	10	40	173	259	19	167	296	4928
Myanmar	24	58	116	466	12	18	1	11	181	3921
Philippines	778	147	293	1173	77	116	8	74	33	1200
Singapore	301	3	0	12	35	33	3	68	21	139
Thailand	2142	252	505	2019	1204	1807	129	1161	230	13980
Vietnam	1733	94	188	751	345	518	37	333	355	3620
Bangladesh	43	3	3	27	12	18	1	12	3	211
Bhutan	9	0	0	1	1	2	0	1	8	12
India	5300	54	54	483	614	922	66	593	4502	29486
Nepal	117	1	1	12	15	23	2	15	98	613
Pakistan	611	9	9	82	90	136	10	87	581	2131
Sri Lanka	276	6	6	58	96	143	10	92	118	839
Asia Total	73970	19249	1722	7017	18478	11298	3024	16885	18106	117577

Table 4. Emission Factors for Fuel Combustion (g kg^{-1} , except for values in shaded area)

Country	Industry		Domestic		Transport		Power	
	Coal	Oil	Coal	Biofuel	Oil		Coal	Oil
SO₂								
China	0.77~117	0.58~11.6	3.29~54.8	0.66~4.12	3.17~10.3		3.81~112.6	3.11~11.6
Japan	0.98~46.9	0.19~15.3	5.57~44.4	0.79~2.52	0.19~16.0		5.57~44.5	0.19~15.3
India	1.32~38.8	9.57~77.5	7.94~15.9	0.18~1.11	9.75~81.3		8.38~16.0	9.75~77.5
Other	5.57~121.7	3.24~72.0	5.28~121.7	0.65~1.53	3.95~25.3		11.7~121.7	19.31~72.0
NO_x								
China	2.38~6.44	3.35~7.26	1.19~2.24	0.54~0.68	15.0~58.2		4.59~11.8	2.1~8.54
Japan	0.51~6.44	3.35~7.26	1.19~2.24	0.54~0.68	15.0~52.0		1.7~11.8	2.1~8.54
India	3.4~6.44	3.35~7.26	1.19~2.24	0.54~0.68	15.0~58.2		4.59~11.8	3.35~8.54
Other	3.4~6.44	3.35~7.26	1.19~2.24	0.54~0.68	15.0~58.2		4.59~11.8	3.35~8.54
CO								
					LDV	HDV	MC	
					(g km^{-1})			
China	37.6		74.0	43~77	1.0~69.3	17.7~135	11.5~21.1	
Japan	1.35		74.0	5.5	2.1~6.8	6.8~40.8	10.0	
India	12.6		74.0	43~77	17.4~60.0	17.4~120	21.0	
Other	37.6		74.0	43~77	12.1~32.7	11.5~40.8	13.0	
BC								
China	0.056~0.6	0.25~0.36	0.12~3.7	1.00	0.01~0.19	0.02~0.27	0.01	0.36
Japan	0.0081~0.06	0.25~0.36	0.12~3.7	1.00	0.003~0.11	0.01~0.16	0.001	0.36
India	0.056~0.6	0.25~0.36	0.12~3.7	1.00	0.01~0.60	0.04~0.87	0.01	0.36
Other	0.030~0.37	0.25~0.36	0.12~3.7	1.00	0.01~0.19	0.02~0.27	0.01	0.36
OC								
China	0.0081~0.056	0.19~0.27	0.12~3.00	5.00	0.01~0.13	0.03~0.18	0.04	0.27
Japan	0.003~0.0081	0.19~0.27	0.12~3.00	5.00	0.006~0.13	0.02~0.18	0.001	0.27
India	0.029~0.056	0.19~0.27	0.12~3.00	5.00	0.05~0.41	0.14~0.59	0.07	0.27
Other	0.018~0.030	0.19~0.27	0.12~3.00	5.00	0.01~0.13	0.03~0.18	0.04	0.27
CO₂								
All	1462~2510	3011~3115	1869~2028	1018~1282	3252		2243~2313	3011~3115

Table 5. Emission Factors for Open Biomass Burning (g kg^{-1})

Species	Savanna/ Grassland	Tropical Forest	Extra-tropical Forest	Crop Residue
SO ₂	0.35	0.57	1.00	0.40
NO _x	3.90	1.60	3.00	2.50
CO ₂	1613	1580	1569	1515
CO	65	104	107	92
CH ₄	2.30	6.80	4.70	2.70
NMVOC	9.73	19.32	27.79	15.70
BC	0.48	0.66	0.56	0.69
OC	3.40	5.20	9.15	3.30
NH ₃	1.05	1.30	1.40	1.30

Table 6. Summary of National Emissions of Each Species in Asia in 2000 (Gg)

Country	SO ₂	NO _x	CO ₂	CO	CH ₄	NMVOC	BC	OC	NH ₃
China	20,385	11,347	3,816,640	115,749	38,356	17,432	1,049	3,385	13,570
Japan	801	2,198	1,202,960	6,806	1,143	1,920	53	74	352
Korea, Rep. of	829	1,322	411,192	2,824	1,433	1,161	22	28	172
Korea, DPR	227	273	120,366	3,556	1,345	234	22	106	98
Mongolia	101	221	68,815	2,861	472	452	19	173	155
Taiwan, China	376	521	199,664	2,127	560	510	8	10	152
Brunei	6	20	10,036	15	50	43	0	0	2
Cambodia	40	89	35,848	1,707	708	305	14	89	86
Indonesia	884	1,317	587,400	23,105	6,443	6,903	206	1,138	1,390
Laos	21	96	43,881	2,547	387	486	18	129	58
Malaysia	273	494	144,071	5,552	869	1,424	26	151	146
Myanmar	65	226	145,315	8,446	2,691	1,671	65	421	341
Philippines	713	326	152,252	4,102	2,563	1,398	36	192	273
Singapore	163	185	55,930	138	85	81	3	2	4
Thailand	961	1,086	350,928	10,815	3,567	3,052	72	364	388
Vietnam	193	283	169,200	9,248	2,907	1,390	88	432	686
Bangladesh	140	220	123,158	4,827	3,608	819	52	268	763
Bhutan	6	8	3,689	172	42	36	2	12	10
India	5,536	4,591	1,886,031	63,340	32,851	10,844	600	2,837	7,399
Nepal	38	55	39,274	2,087	917	346	21	135	168
Pakistan	1,416	539	221,349	7,076	5,415	1,344	85	368	1,214
Sri Lanka	58	57	28,309	1,348	407	275	11	55	92
International Shipping	1,083	1,292	51,280	117	1	27	68	51	0
Asia Total	34,316	26,768	9,867,589	278,564	106,821	52,150	2,541	10,420	27,519

Table 7. Speciation of NMVOC by Country in Asia in 2000 (Gg)

Country	Alkanes	Alkenes	Ethyne	Aromatics	Ald./ket.	Other	Total ^a
China	5,067	3,629	892	3,435	1,324	3,094	17,440
Japan	611	141	20	301	83	770	1,925
Korea, Rep. of	507	130	25	201	48	253	1,164
Korea, DPR	87	27	6	55	15	44	234
Mongolia	36	64	10	29	108	205	452
Taiwan, China	257	39	7	88	21	100	512
Brunei	38	1	0	2	0	3	43
Cambodia	48	67	12	34	53	91	305
Indonesia	1,577	1,745	338	1,250	614	1,395	6,918
Laos	58	95	13	34	111	176	486
Malaysia	485	239	39	218	141	304	1,426
Myanmar	256	354	54	174	318	516	1,672
Philippines	279	292	59	225	157	393	1,404
Singapore	24	9	1	10	9	28	81
Thailand	783	618	108	565	312	670	3,056
Vietnam	275	375	72	214	167	287	1,391
Bangladesh	164	208	39	98	113	197	820
Bhutan	7	8	1	5	5	10	36
India	2,826	2,811	588	1,796	955	1,874	10,850
Nepal	53	96	19	49	45	85	346
Pakistan	416	331	71	214	101	211	1,344
Sri Lanka	68	67	12	45	28	55	275
International Shipping	1	20	3	2	0	0	27
Asia Total	13,920	11,365	2,390	9,044	4,729	10,759	52,206

^aRounding errors in the speciation calculations cause these numbers to be slightly different to those in Table 6.

Table 8. Uncertainty (%) in Pollutant Emission Estimates ($\pm 95\%$ Confidence Intervals)

Region	SO ₂	NO _x	CO ₂	CO	CH ₄	NMVOC	BC	OC	NH ₃
China	13	23	16	156	71	59	484	495	53
Japan	9	19	7	34	52	35	83	181	29
Other East Asia	12	24	13	84	101	49	160	233	31
Southeast Asia	27	92	91	214	95	218	257	345	87
India	26	48	33	238	67	149	359	544	101
Other South Asia	35	63	44	291	109	148	379	531	101
International Shipping	44	56	40	72	72	204	402	402	—
All Asia	16	37	31	185	65	130	364	450	72

Table 9. Comparison of Observed and STEM-Simulated Mean Concentrations for TRACE-P DC-8 Flights #6 through #17 at Altitudes Below 1 km

Species (units)	Observed	Modeled
CO (ppbv)	219	203
Ethane (ppbv)	2.0	1.6
Propane (ppbv)	0.62	0.46
Ethene (ppbv)	0.18	0.20
Ethyne (ppbv)	0.78	0.63
SO ₂ (ppbv)	1.55	1.04
NO ₂ (ppbv)	0.27	0.25
NO (ppbv)	0.035	0.041
Benzene + toluene (ppbv)	0.33	0.19
BC (μg m ⁻³)	0.84	0.67

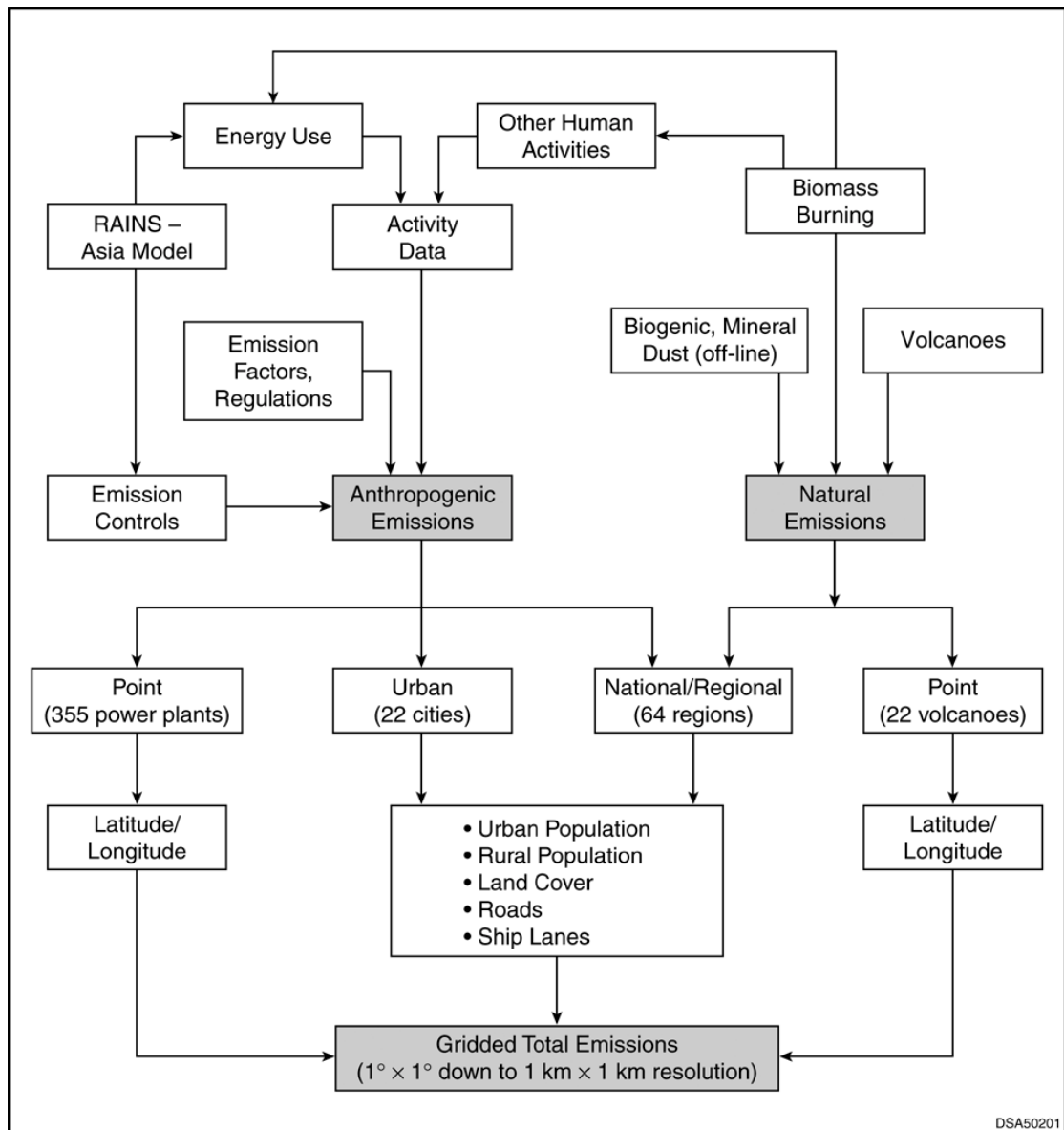
Table 10. Asian Emission Trends (Gg) from Anthropogenic Sources (No Biomass Burning Included)

Species	Region	1990	1994	1995	2000	94-00 (%)	Brief Explanation
SO ₂ (Tg)	China	22.2 ^a	24.2 ^a	25.7 ^a	20.3 ^b	-16.1	reduction in coal use; switch to lower-S coals
	East Asia ^c	25.9 ^a	27.3 ^a	27.9 ^a	22.6 ^b	-17.2	China dominates S; lower-S oil in Taiwan and ROK
NO _x (Tg)	China	7.2 ^d	9.2 ^e	9.7 ^d	10.3 ^b	13.0	growth in transport offsets decline in coal use
	East Asia ^c	10.8 ^d	13.3 ^e	13.9 ^d	14.8 ^b	12.0	transport growing slower in developed East Asia
NMVOC (Tg)	China	11.4 ^d	12.8 ^e	13.1 ^d	14.6 ^b	14.1	unabated growth in chemicals, solvents, oil use
	East Asia ^c	15.2 ^d	16.7 ^e	17.1 ^d	18.5 ^b	10.8	emissions better controlled in developed East Asia
NH ₃ (Tg)	China	9.7 ^d	11.3 ^e	11.7 ^d	13.3 ^b	17.7	increase in fertilizer use in recent years
	East Asia ^c	10.6 ^d	12.2 ^e	12.6 ^d	14.2 ^b	16.4	China dominates NH ₃ emissions in East Asia
CO ₂ ^f (Pg)	China	30.7 ^g	35.9 ^g	37.1 ^g	36.3 ^g	1.1	reduction in coal use, improved energy efficiency
CH ₄ (Tg)	China	29.1 ^g	29.8 ^g	30.6 ^g	31.7 ^g	6.4	reduced coal mining has slowed increase
BC (Tg)	China	N.A.	N.A.	1.27 ^h	0.94 ^b	-26.0	reduction in coal use by households and industry

^a*Streets et al.*, 2000a.^bThis work.^cIncludes China; Japan; DPRK; ROK; Mongolia; and Taiwan, China.^d*Klimont et al.*, 2001.^eLinearly interpolated between 1990 and 1995 values (growth was approximately linear in this period.)^fDirect emissions only; does not include take-up by growing vegetation.^g*Streets et al.*, 2001b.^h*Streets et al.*, 2001c; the trend shown for BC is from 1995-2000 (no 1994 value available).



Figure 1



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Figure 2

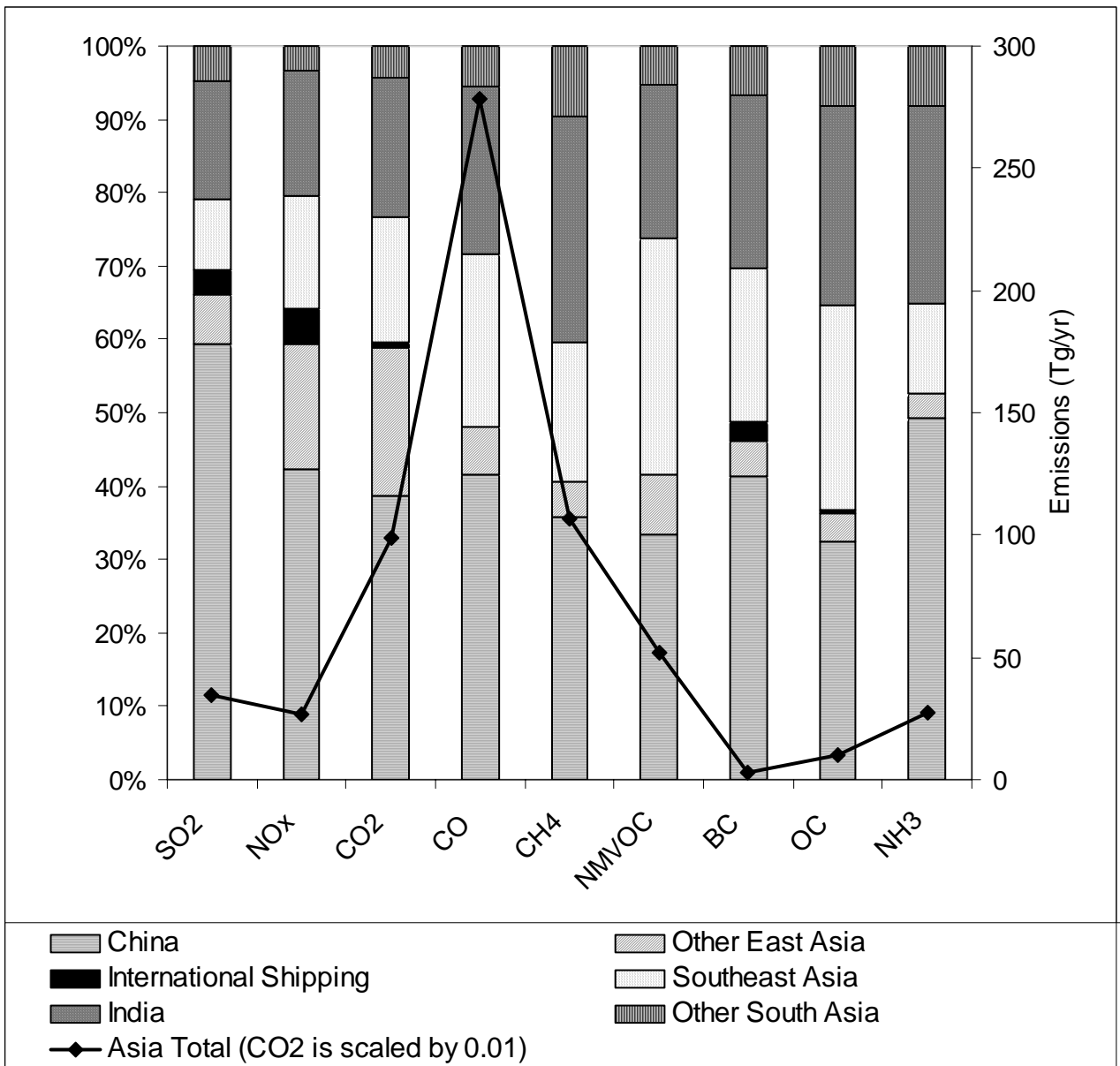


Figure 3

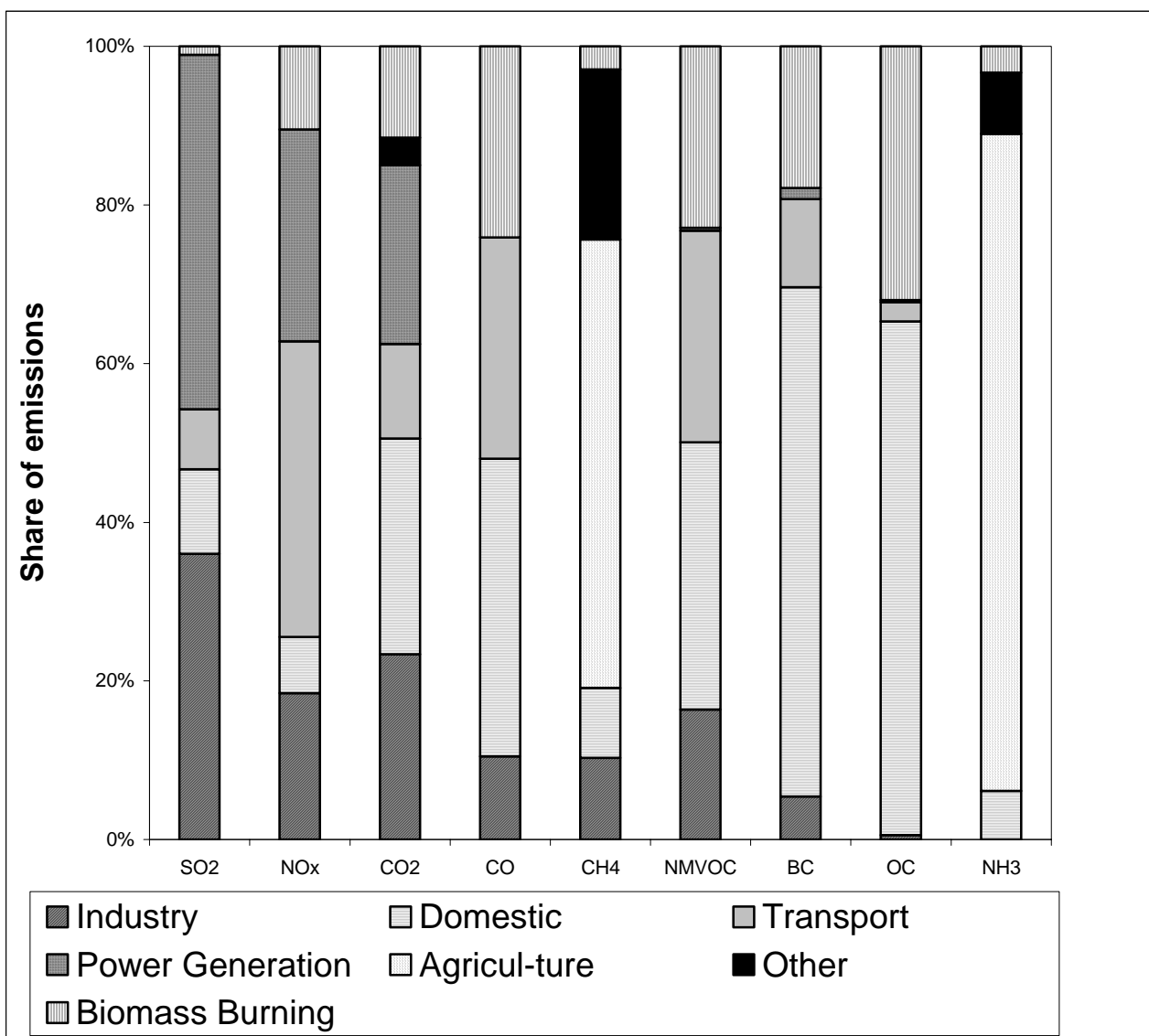


Figure 4

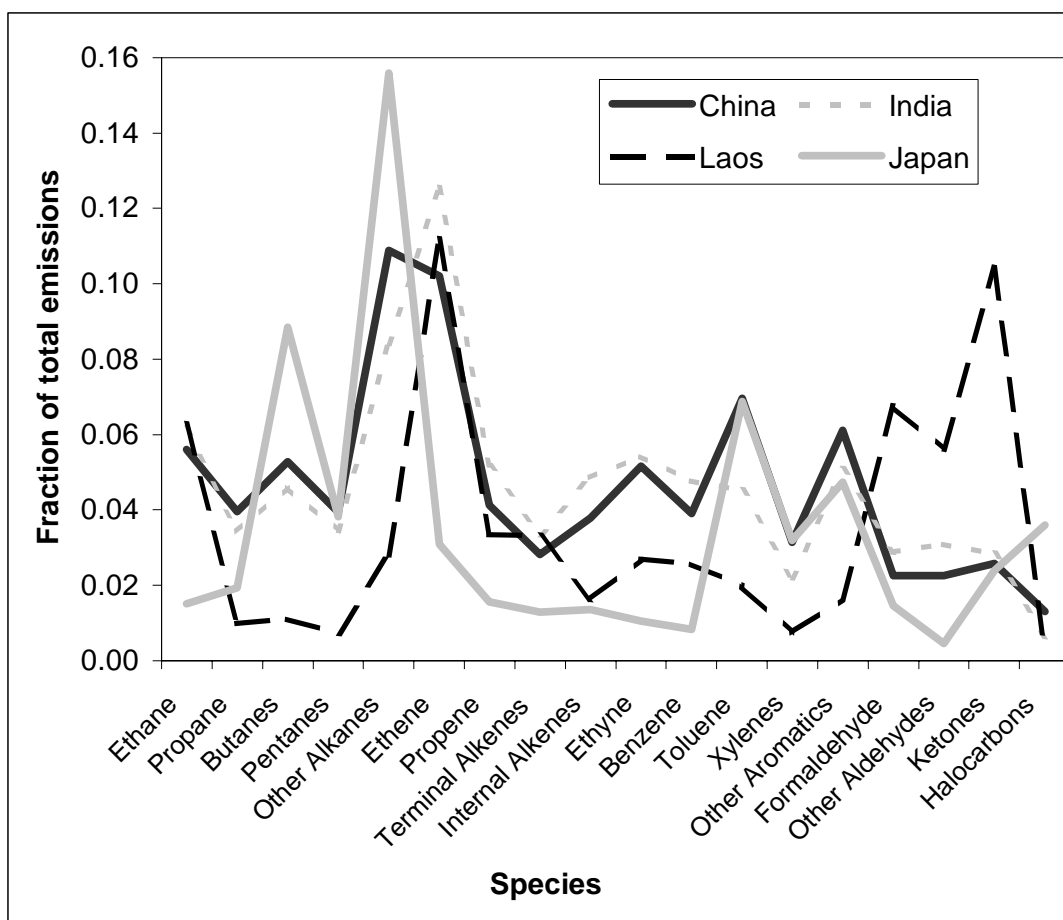


Figure 5

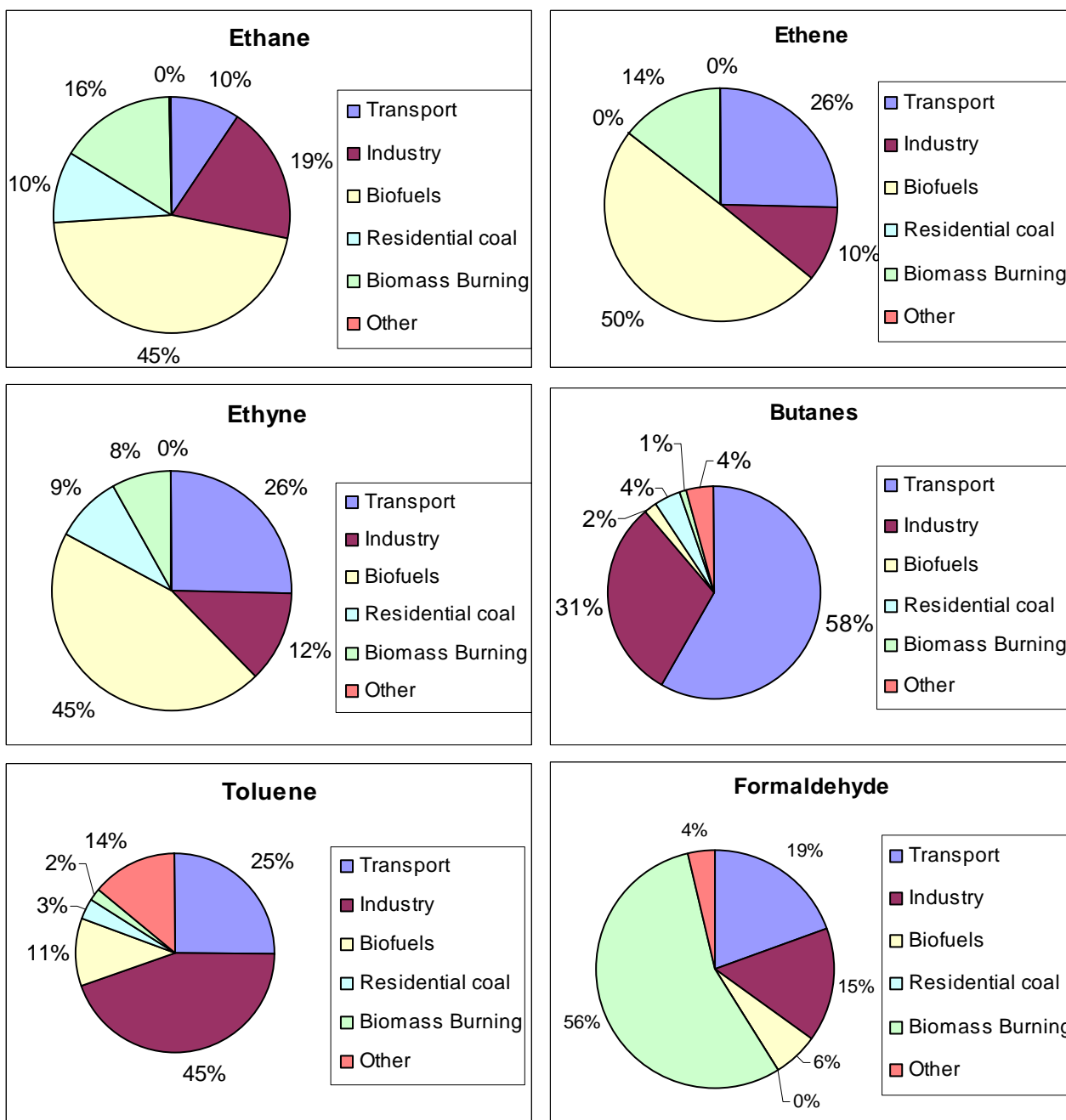


FIGURE 6

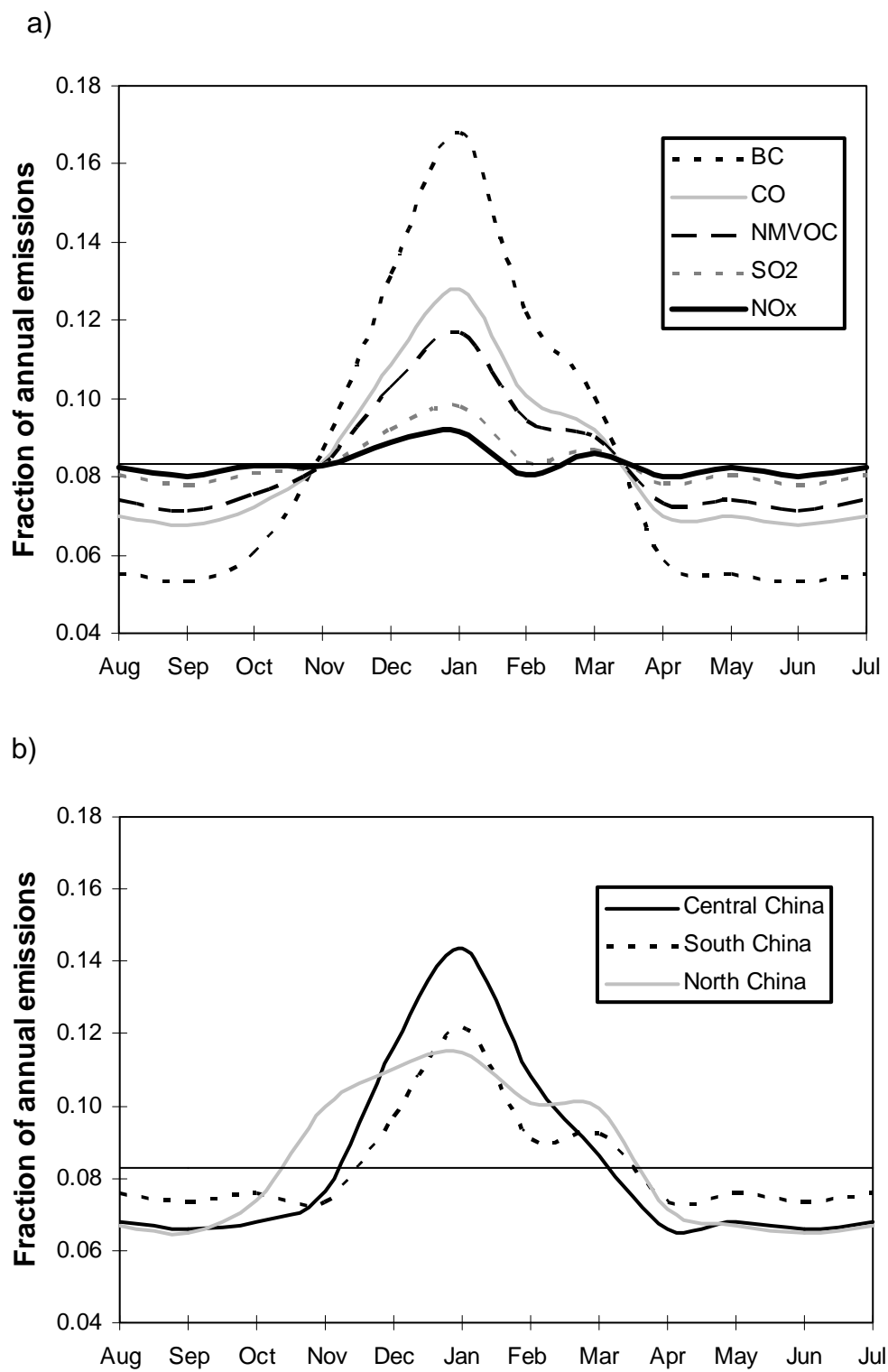


Figure 7

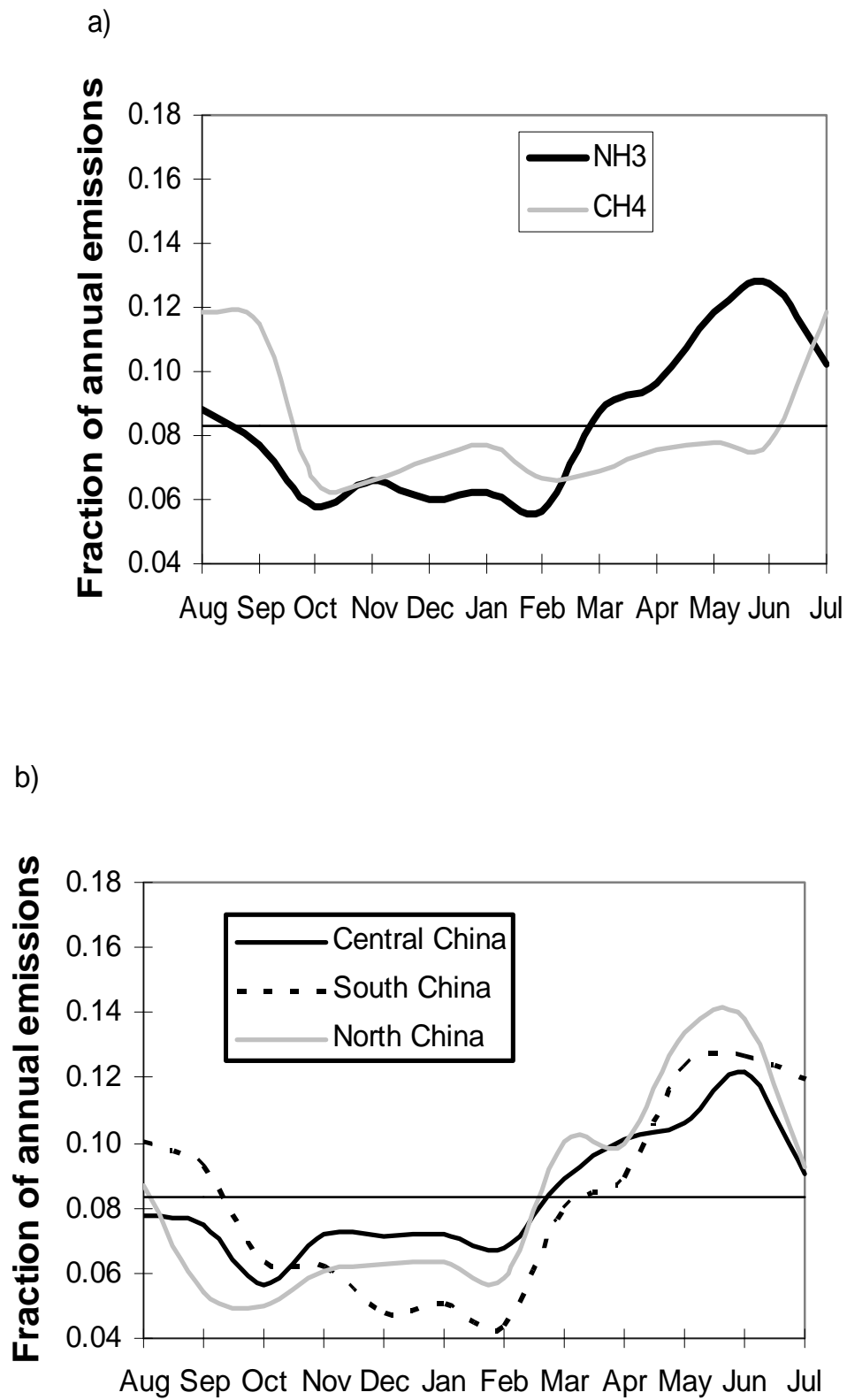


Figure 8

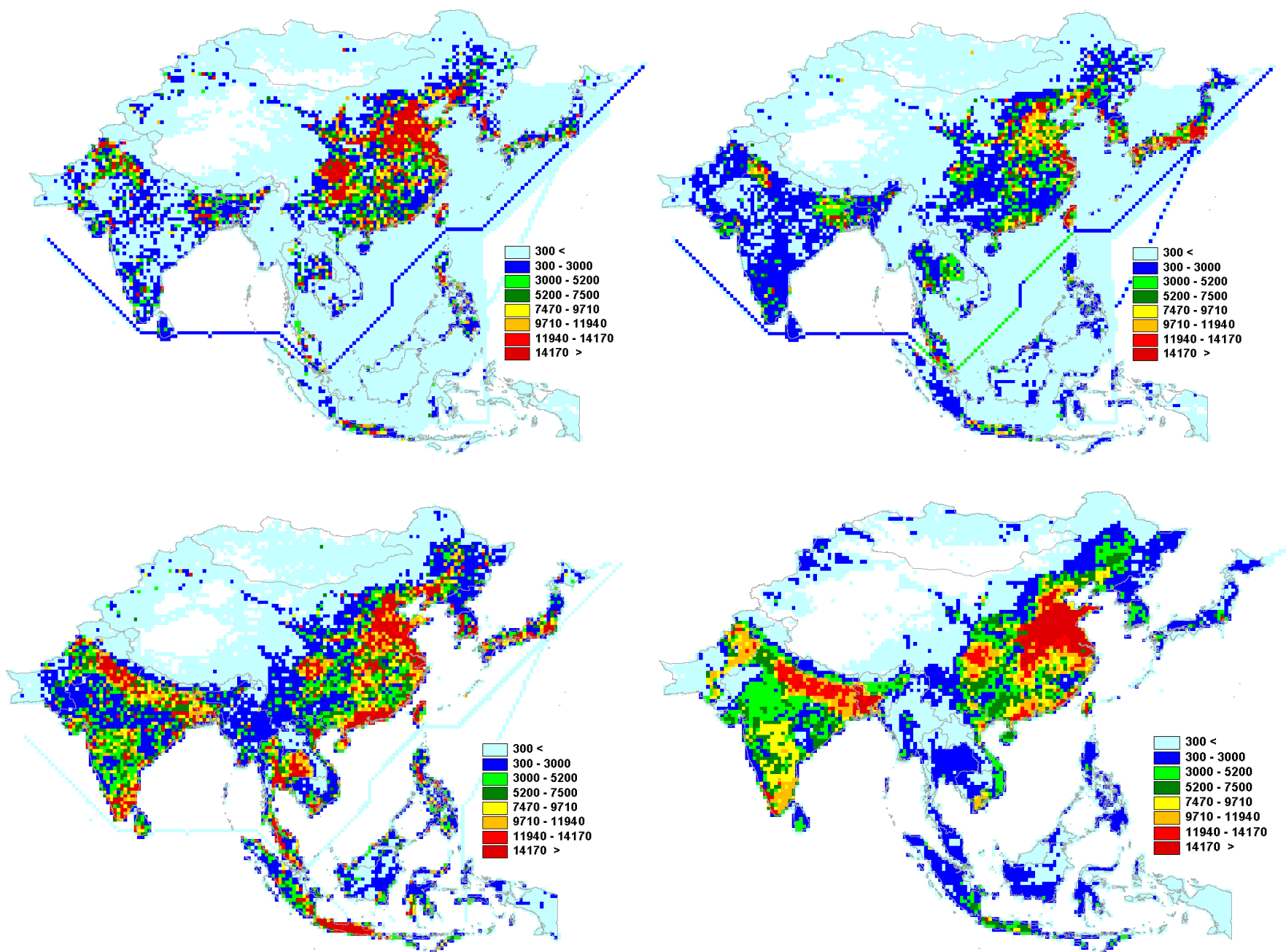


Figure 9

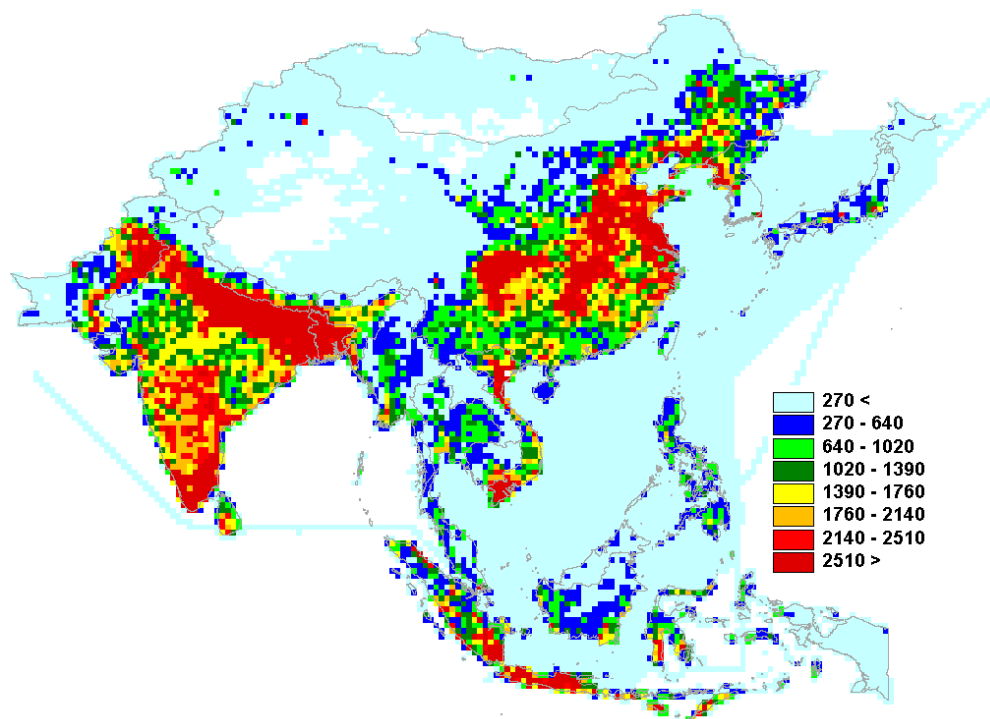
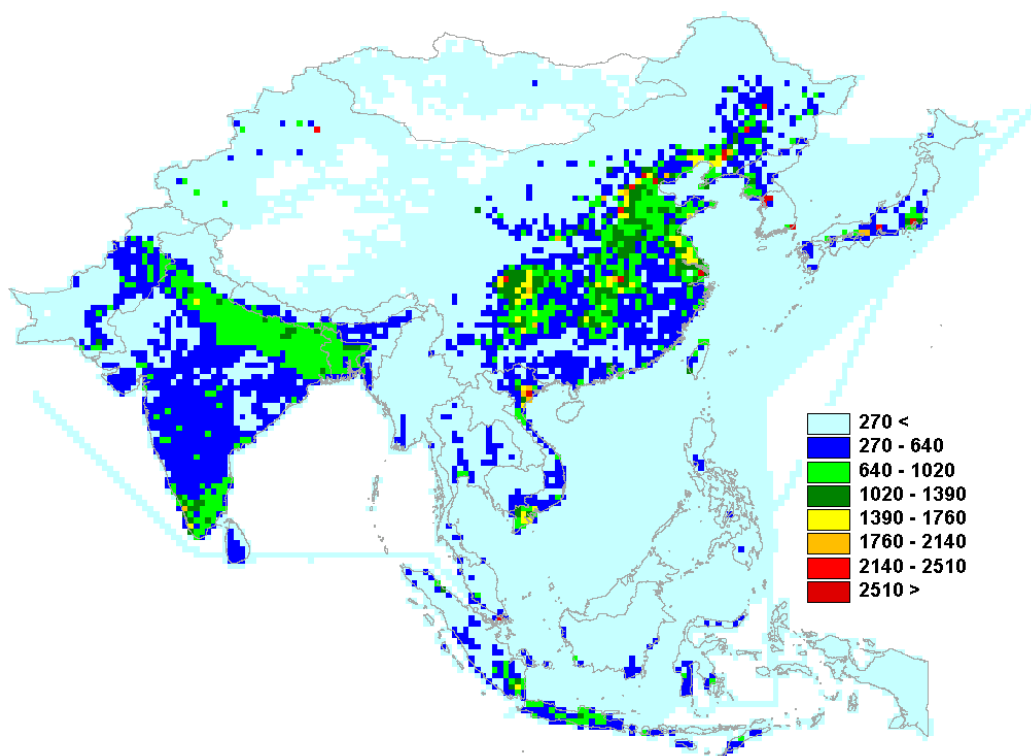


Figure 10

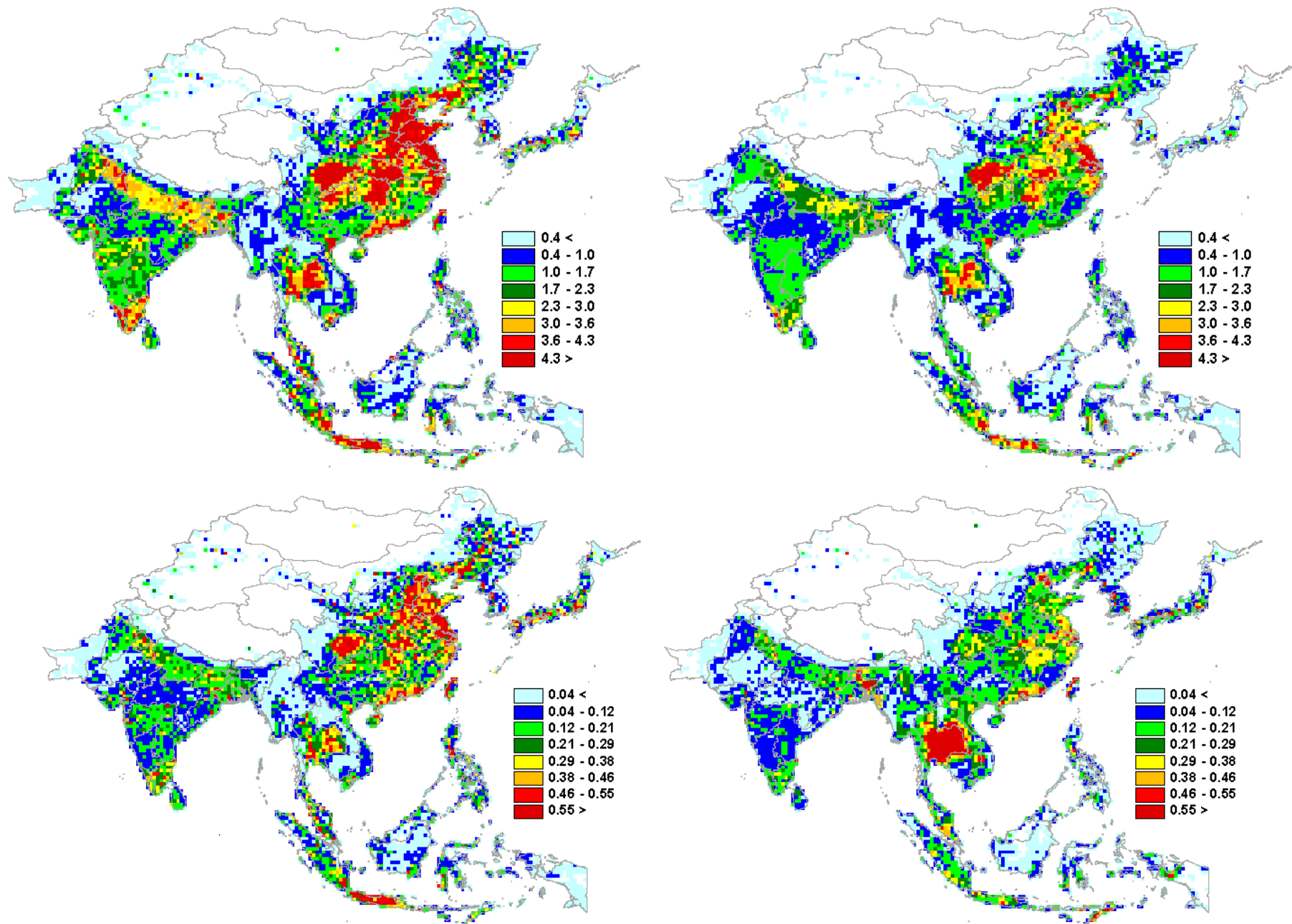


Figure 11

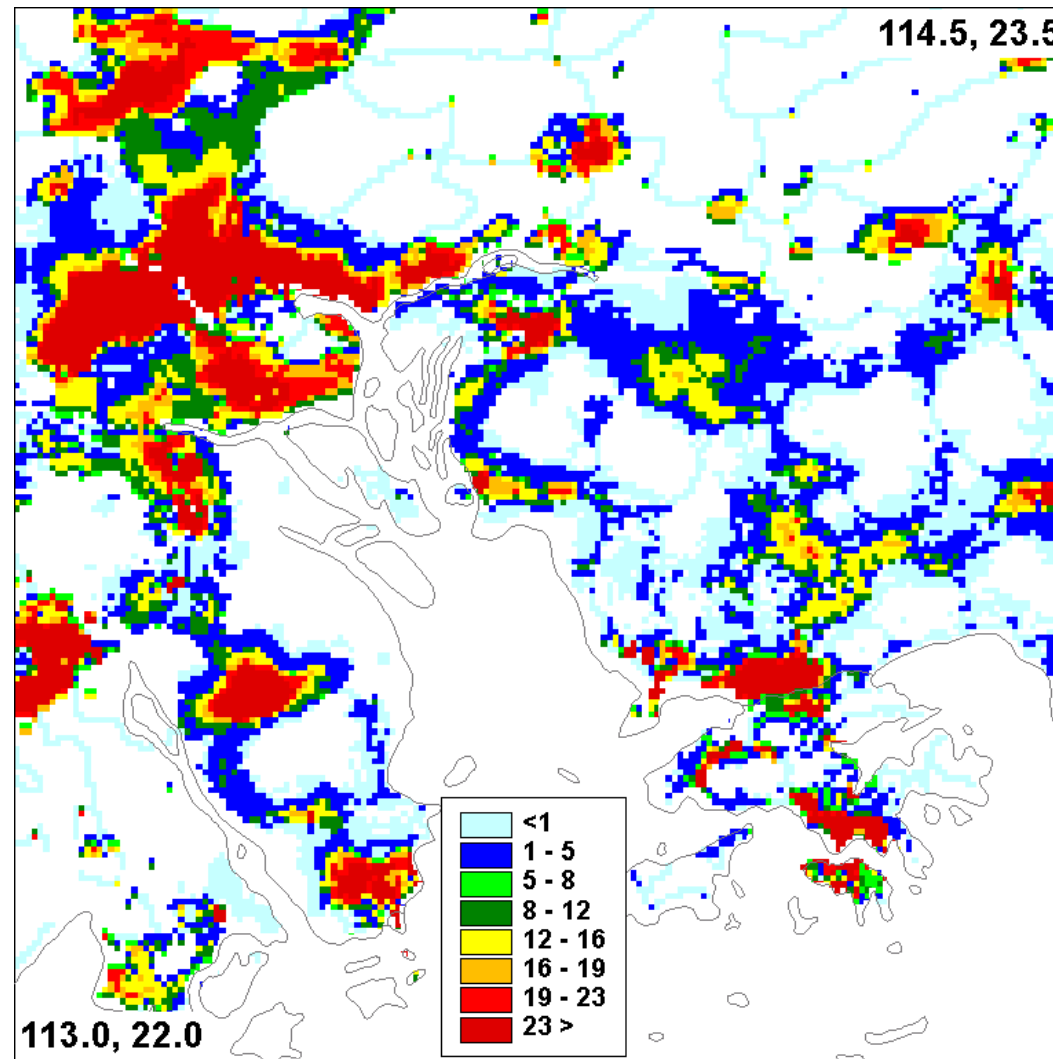


FIGURE 12 (a)

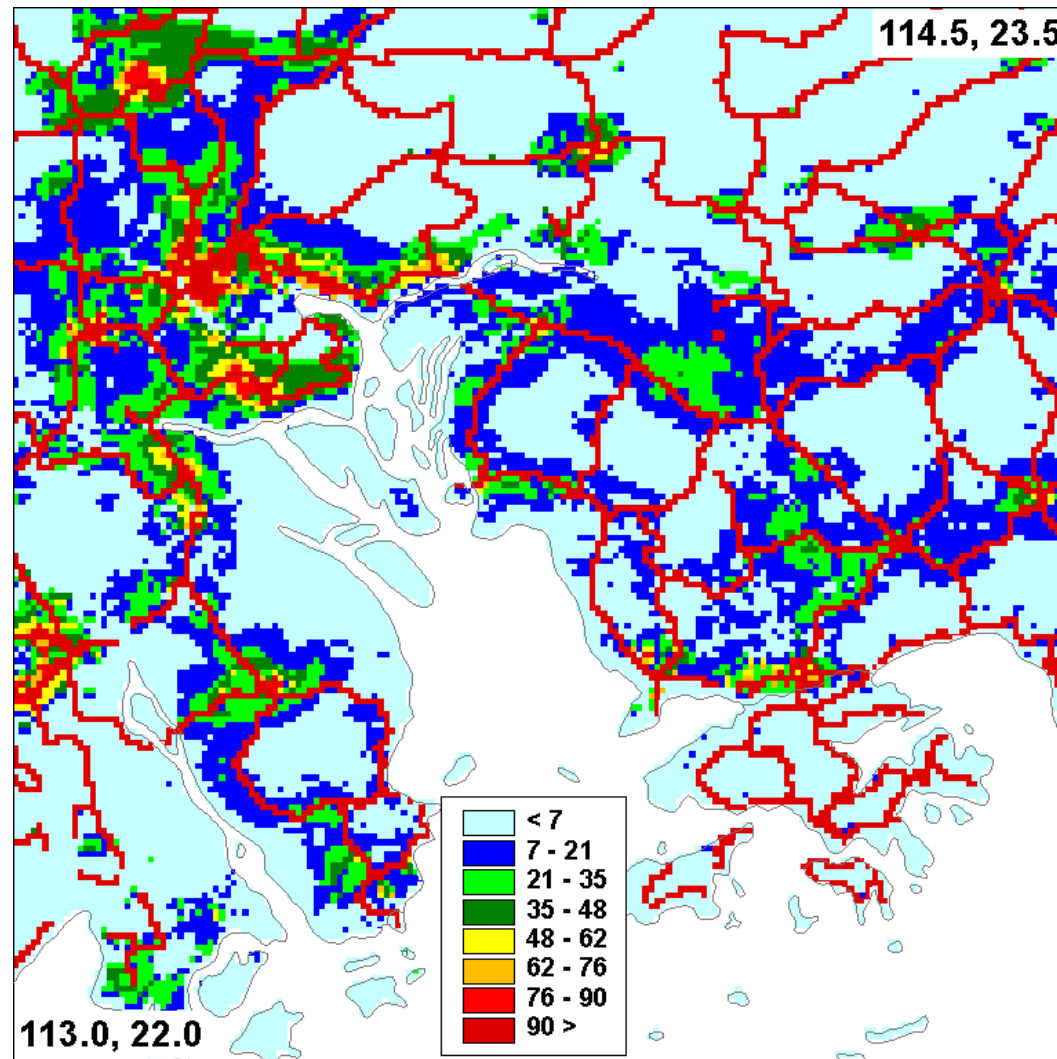


FIGURE 12 (b)

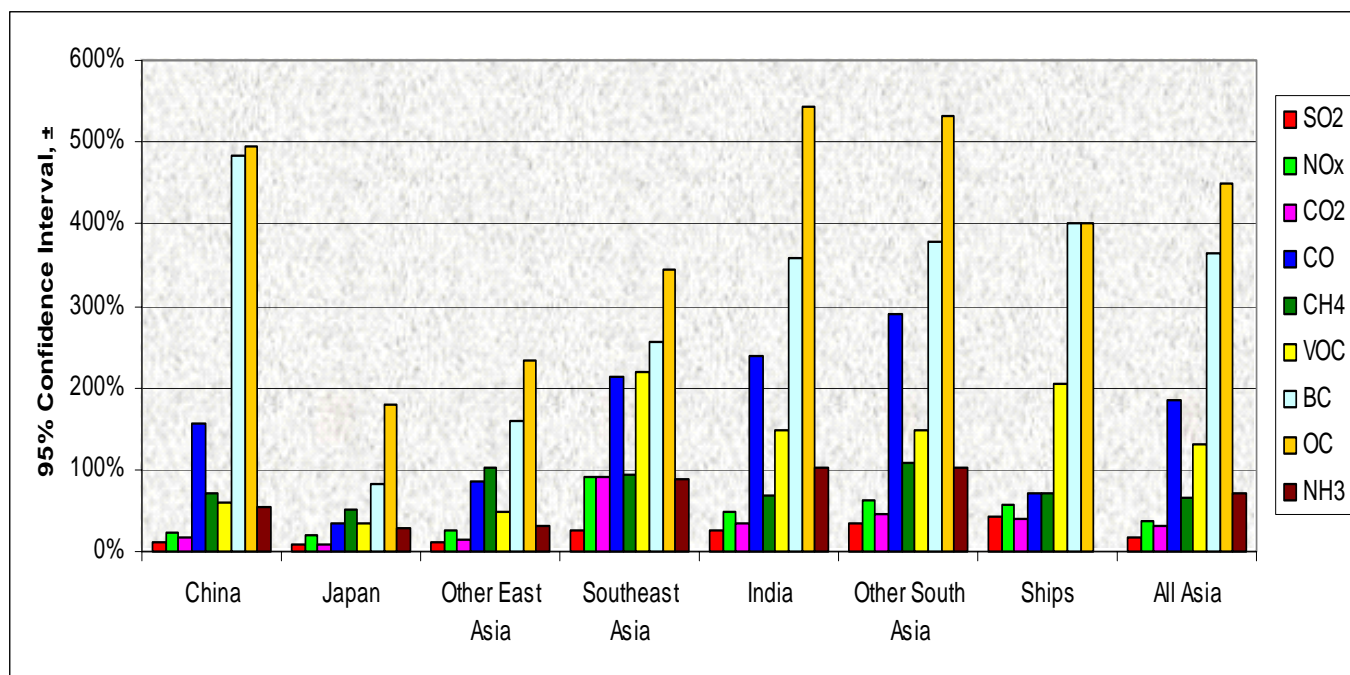
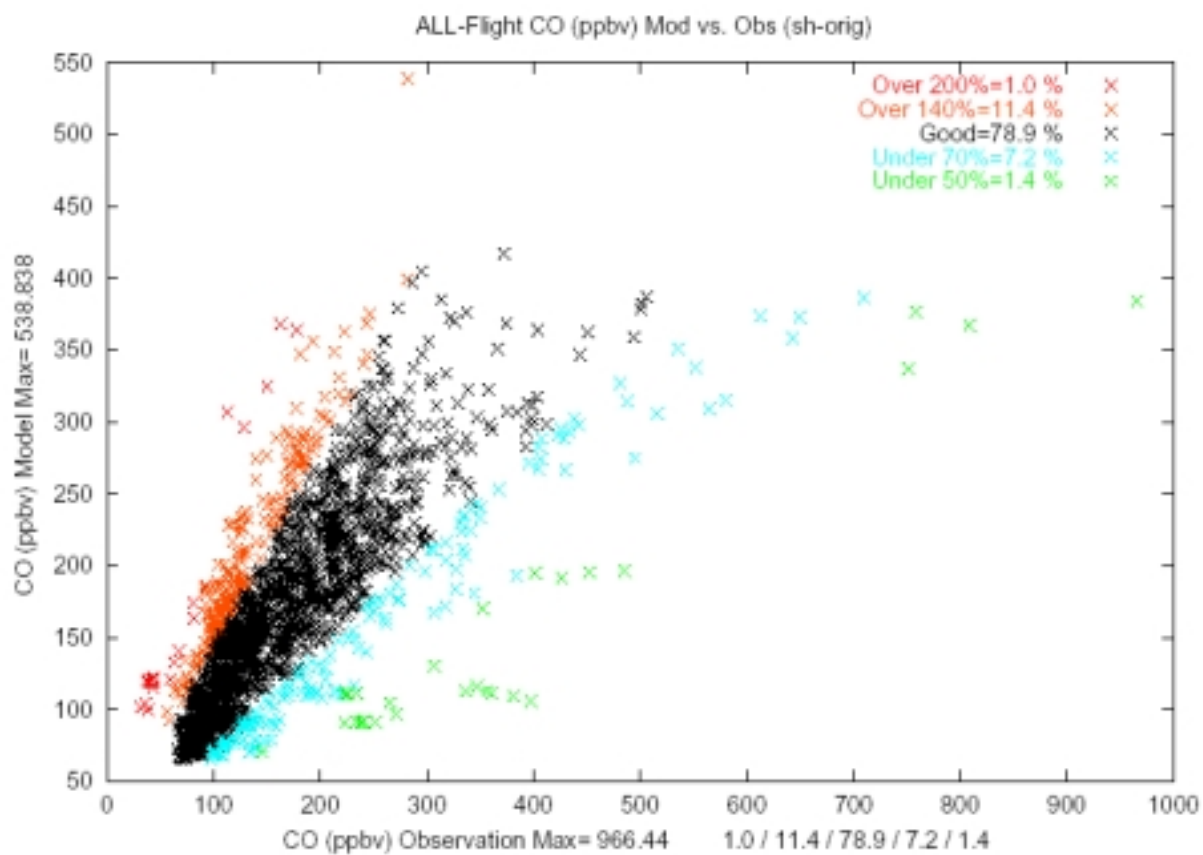


Figure 13

**Figure 14**